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# PRE-HISTORY OF THE CONCEPTS UNDERLYING STIMULATED RAMAN ADIABATIC PASSAGE (STIRAP)

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This tutorial review discusses some of the work that preceded development, twenty-five years ago, of the stimulated Raman adiabatic passage (STIRAP) technique, now widely used in the controlled coherent dynamics of three-state systems, noting how the use of time-dependent adiabatically-evolving population-trapping dark states made possible the robust and highly-efficient population transfer between quantum states that first popularized STIRAP. Preceding the history discussion is a tutorial definition of STIRAP and its necessary and sufficient ingredients — understanding that has led to applications well beyond those of the original quantum systems. This review also discusses the relationship between STIRAP and two related procedures: chirped Raman adiabatic passage (RCAP or CHIRAP) and electromagnetically induced transparency (EIT) with slow and captured light. It concludes with a brief discussion of ways in which contemporary STIRAP has extended the original concept and enlarged the definition, beyond that of simple quantum systems to classical macroscopic devices. Appendices offer further details. The presentation emphasizes theory but with illustrations of experimental results.

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## Contents



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## 1 Introduction: STIRAP defined

The technique of coherent manipulation of quantum states known as stimulated Raman adiabatic passage (STIRAP), originally developed as a procedure for efficiently producing population transfer between vibrational levels of molecules in beams [Gau88, Kuk89, Gau90], has been broadly extended in a variety of ways in the subsequent years to systems and applications differing substantially from those of the initial interest. Several reviews [Ber95, Sho95b, Ber98, Vit01a, Vit01b] and monograph sections [Sho08, Sho11] have, from time to time, provided detailed description of the underlying concepts and overviews of its many applications and experimental demonstrations.

This present article helps celebrate the quarter century of STIRAP existence by looking back upon earlier works, preceding but related to aspects of STIRAP, pointing out both what they did and what they did not do. Here is an outline of the contents:

**Section** 1.1 describes the underlying Raman and stimulated-Raman scattering (SRS) processes.

**Section** 1.2 provides a working definition of the STIRAP procedure and the equations associated with it.

**Section** 1.3 discusses the several identifying characteristics associated with the STIRAP process.

**Section** 2 reviews a number of earlier works, ordered roughly by topic and date of publication, whose results form parts of the overall procedure that is here called STIRAP. Many of these made some notable contribution to the theory[2], but none were a complete example of STIRAP.

**Section** 3 continues the historical record to summarize the first development of STIRAP .

**Section** 4 discusses the relationship of STIRAP to an earlier procedure, known as RCAP or CHIRAP, for accomplishing adiabatic transfer of population using frequency sweeping (chirping) in a stimulated-Raman linkage.

**Section** 5 discusses some procedures developed at the time of STIRAP dealing with manipulations of *fields*, rather than *quantum states*, and that have close connections with STIRAP: Electromagnetically induced transparency (EIT), slow light and the trapping and retrieval of light pulses.

**Section** 6 summarizes some of the extensions to the original paper on STIRAP that currently provide a somewhat enlarged definition of STIRAP, extending beyond the stimulated Raman process of molecular excitation to find application in many research areas, including macroscopic classical systems.

**Section** 7 presents a brief general summary.

**Appendices** present comments on notation and nomenclature and supplementary matters related to coherent excitation, dark states and EIT.

[2]The story of earlier experiments must be told elsewhere.

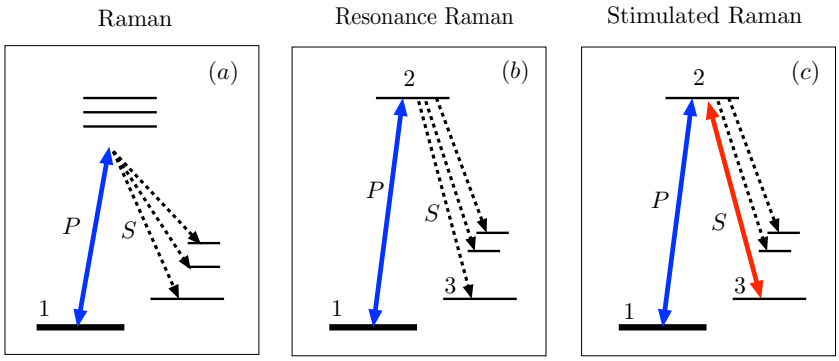


Fig. 1.1. $(a)$ The two-photon Raman process: a primary (pump) field $P$ is accompanied by a set of spontaneously-emitted secondary $S$ fields (dashed lines). Often selection rules make a single spontaneous-emission field dominant. $(b)$ The resonance Raman process: a field $P$ links the ground state 1 with an excited state 2 which decays by spontaneous emission (dashed lines) to state 3 as well as other states. The frequencies satisfy the two-photon resonance condition (1.3). $(c)$ The stimulated Raman scattering (SRS) process: a specific $S$ field links intermediate state 2 with a specific final state 3. Spontaneous emission occurs as well. In each frame the heavy horizontal line for state 1 emphasizes that this is initially populated.

### 1.1 Raman and stimulated Raman processes; Double optical resonance

The basic traditional *Raman process* [Ram28a, Ram28b, Ram28c, Ram28d, Bor28b], long used in molecular spectroscopy [Her50, Col90, Fer03, Smi05], is an inelastic light-scattering process in which a photon from a *primary* (or pump) field $P$ field is scattered into a *secondary* (or Stokes) field $S$, with the difference in photon energies going to an energy change of a molecule, most commonly rotational or vibrational excitation. A single $P$ field of given frequency will produce a set of $S$ fields, whose measured frequencies allow determination of the excitation energies of the material. Frame $(a)$ of **Fig.** 1.1 illustrates this two-photon process.

**Resonance Raman.** In the more specialized *resonance Raman* scattering (RRS) processes, shown in frame $(b)$ of **Fig.** 1.1, the excitation step uses a $P$ field that brings population from an initially populated state 1, of energy $E_1$, into a specific excited electronic state 2, of energy $E_2$, by having a carrier frequency $\omega_P$ that is very close to satisfying the *single-photon resonance* condition with the 1-2 transition,

$$\hbar\omega_P = |E_2 - E_1|. \tag{1.1}$$

There follows population transfer into stable or metastable states by spontaneous emission of $S$-field photons. For a given final state 3, of energy $E_3$, the $S$-field frequency $\omega_S$ is set by the 2-3 transition energy,

$$\hbar\omega_S = |E_2 - E_3|. \tag{1.2}$$

In the present article "Raman" will generally refer to the resonance (or near-resonant) Raman process, via a single intermediate state. The (non-resonant) Raman process of **Fig.** 1.1$(a)$ proceeds

through numerous virtual intermediate states which are all far from single-photon resonance.

**Stimulated Raman.** The *stimulated* Raman process (or stimulated Raman scattering, SRS) [Eck62, Hel63, Zub64, She65, Blo67, Wan69, Cou77, Ray79, Ray81, Ray90] shown in **Fig.** 1.1$(c)$, replaces the spontaneously emitted $S$ fields by a controlled radiation field: the 2-3 transition is, in the Einstein view (see **App.** D), a *stimulated emission*. In general for $SRS$ the intermediate state 2 need not be resonant with either $P$ or $S$ alone (as is shown here), but the difference of those two frequencies must equal the 1-3 transition frequency: they satisfy the *two-photon resonance* condition (see **Fig.** 1.2 for linkage display):

$$|E_3 - E_1| = \begin{cases} \hbar|\omega_P - \omega_S|, & E_1, E_3 < E_2, \quad \textit{lambda linkage,} \\ \hbar(\omega_P + \omega_S), & E_1, < E_2 < E_3, \quad \textit{ladder linkage.} \end{cases} \tag{1.3}$$

The spontaneous-emission fields will always accompany the $S$ field, being generated at a fixed rate, the inverse of the spontaneous-emission lifetime. Their presence can be ignored during the excitation stage when the $S$ field is sufficiently brief and strong. They are detectable as scattering or fluorescence.

**Stokes and anti-Stokes.** When $E_3 > E_1$ (and hence $\omega_P > \omega_S$), as shown in **Fig.** 1.1, state 3 is an excited state and the secondary $S$-radiation is termed *Stokes* radiation. When $E_1 > E_3$ (and hence $\omega_P < \omega_S$), state 1 is an excited state — the SRS takes energy *away from* the molecule — and the $S$ radiation is historically termed *anti-Stokes* radiation, a distinction that will not be followed in the present article: the SRS and STIRAP processes do not depend on the relative values of $E_1$ and $E_3$.

## 1.2 Stimulated Raman adiabatic passage (STIRAP)

**The linkage.** STIRAP is one of many procedures that use pulses of coherent radiation to induce population transfer between two quantum states [Sho90, Sho11]. The prototypical quantum system in which STIRAP occurs comprises three quantum states, 1,2,3, linked in a chain by two monochromatic radiation fields $P$ and $S$ (the double-headed arrows signify that the interactions are coherent, moving probabilities both ways):

$$1 \underset{P}{\longleftrightarrow} 2 \underset{S}{\longleftrightarrow} 3. \tag{1.4}$$

The three-state chain is common to many physical systems, some of which will be discussed here. The linkage pattern of the Hamiltonian energy matrix, viewed as a *graph* [Ein76, Ein79], has three *nodes* (quantum states or Zeeman sublevels, 1,2,3) and two *links* (fields or Rabi frequencies $P, S$). Linkage patterns are also known as *configurations*.

### 1.2.1 The equations of motion

**The statevector.** Descriptions of coherent manipulation of discrete quantum states rely on a time-dependent statevector $\mathbf{\Psi}(t)$ defined in an $N$-dimensional Hilbert space whose coordinate

unit-vectors $\psi_n$ are associated with the $N$ basic distinguishable quantum states of interest; cf. **App.** B.1. The general statevector construction reads:

$$\mathbf{\Psi}(t) = \sum_{n=1,N} C_n(t)\ \exp[-\mathrm{i}\zeta_n(t)]\,\boldsymbol{\psi}_n. \tag{1.5}$$

Mathematically the coefficients $C_n(t)$ are expansion coefficients in a system of coordinates $\boldsymbol{\psi}'_n(t)$ that rotate with the phases $\zeta_n(t)$, see **App.** C:

$$\boldsymbol{\psi}'_n(t) \equiv \exp[-\mathrm{i}\zeta_n(t)]\,\boldsymbol{\psi}_n. \tag{1.6}$$

It is these (rotating) vectors that serve as the unit column-vectors in the present discussion (and, implicitly, in almost all other papers discussed here):

$$\begin{bmatrix} 1 \\ 0 \\ 0 \end{bmatrix} = \boldsymbol{\psi}'_1(t), \qquad \begin{bmatrix} 0 \\ 1 \\ 0 \end{bmatrix} = \boldsymbol{\psi}'_2(t), \qquad \begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix} = \boldsymbol{\psi}'_3(t). \tag{1.7}$$

Physically the $C_n(t)$ are *probability amplitudes* for the probability

$$P_n(t) = |C_n(t)|^2 \tag{1.8}$$

of finding the system to be in state $n$ at time $t$ — the instantaneous *population* of state $n$.

**The TDSE.** The construction (1.5) is used with the time-dependent Schrödinger equation (TDSE) and Hamiltonian operator $\mathsf{H}(t)$,

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\mathbf{\Psi}(t) = \mathsf{H}(t)\mathbf{\Psi}(t), \tag{1.9}$$

to produce a set of coupled ordinary differential equations (ODEs), as implementation of the TDSE:

$$\mathrm{i}\frac{d}{dt}\mathbf{C}(t) = \mathsf{W}(t)\mathbf{C}(t), \tag{1.10}$$

where $\mathbf{C}(t)$ is a column-vector having probability amplitudes $C_n(t)$ as elements and the elements of the matrix $\mathsf{W}(t)$ have units of angular frequency, i.e. radians per unit time.

Whereas the Hamiltonian $\mathsf{H}(t)$ typically contains terms that oscillate at optical frequencies, the population changes of interest occur more slowly (an example of multiple time-scale perturbation-theory [Won76, Won77]). The appropriate choice of phases $\zeta_n(t)$, together with neglect of rapidly varying terms — *the rotating-wave approximation* (RWA), cf. **Sec.** 1.3.2 and [Sho90, Sho11] — typically makes the coefficient matrix $\mathsf{W}(t)$ only slowly varying or constant.

The Hamiltonian matrix[3] $\mathsf{W}(t)$ for STIRAP is customarily presented in the rotating-wave picture (see **App.** C) using the rotating-wave approximation. In the present article I generally

[3]Some authors use units such that $\hbar = 1$, blurring the distinction between angular frequency and energy, and making more obvious the referral to both $\mathsf{W}(t)$ and $\mathsf{H}(t)$ as Hamiltonian matrices.

choose the phases $\zeta_n(t)$ and the state orderings such that the 1,1 element of $\mathsf{W}(t)$ is zero [using eqn. (C.4a) of **App.** C]. With two-photon resonance as required for STIRAP [cf. eqns. (1.3) and (1.18)] and appropriately chosen phases the constructions read :

$$\mathsf{W}(t) \equiv \frac{1}{2}\begin{bmatrix} 0 & \Omega_P(t) & 0 \\ \Omega_P(t) & 2\Delta & \Omega_S(t) \\ 0 & \Omega_S(t) & 0 \end{bmatrix}, \qquad \mathbf{C}(t) \equiv \begin{bmatrix} C_1(t) \\ C_2(t) \\ C_3(t) \end{bmatrix}. \tag{1.11}$$

These three coupled ordinary differential equations (ODEs) for the three complex-valued probability amplitudes $C_n(t)$ are the fundamental equations of STIRAP (see **Sec.** 6.8). They involve three parameters defined in the paragraphs below: two time-varying *Rabi frequencies* $\Omega_P(t)$ and $\Omega_S(t)$ and a constant *detuning* $\Delta$, possibly null.

**Rabi frequencies.** The two off-diagonal elements of $2\mathsf{W}(t)$, denoted $\Omega_P(t)$ and $\Omega_S(t)$ and responsible for transitions 1-2 and 2-3 respectively, are (time-dependent) Rabi frequencies[4] [Rab54, Kni80, Sho90]. This article takes them to be real-valued and slowly varying. They parametrize interaction energies, and may have as their origin an electric-dipole interaction, as is common in quantum optics, or a magnetic-dipole interaction, as is used in magnetic resonance work. They may also express induced-dipole multiphoton transitions, as in hyper-Raman adiabatic passage [Yat98b]; see **Sec.** 2.10.1. But, as noted in **Sec.** 6.8, they may also have a very different, quasi-static origin.

**The interaction and fields.** The portion of the Hamiltonian responsible for most optical transitions, including stimulated Raman scattering, is that of an electric dipole moment $\mathbf{d}$ in an electric field $\mathbf{E}(t)$, evaluated at the center of mass for the system,

$$\mathsf{H}^{\rm int}(t) = -\mathbf{d} \cdot \mathbf{E}(t), \tag{1.12}$$

but other interactions have interest [Sho90, Sho11] and this form is not essential for discussions of coherent excitation. In the original STIRAP procedure the two pulsed fields have constant carrier frequencies $\omega_P$ and $\omega_S$. That is, the electric field producing the interaction (1.12) has the form

$$\mathbf{E}(t) = \mathrm{Re}\Big[\, \mathbf{e}_P(t)\, \mathcal{E}_P(t)\, \exp(-\mathrm{i}\omega_P t + \mathrm{i}\varphi_P) + \mathbf{e}_S(t)\, \mathcal{E}_S(t)\, \exp(-\mathrm{i}\omega_S t + \mathrm{i}\varphi_S)\Big], \tag{1.13}$$

where $\mathbf{e}_P(t)$ and $\mathbf{e}_S(t)$ are unit vectors, either time independent or slowly varying, $\mathcal{E}_P(t)$ and $\mathcal{E}_S(t)$ are slowly-varying pulse envelopes[5], and $\varphi_P$ and $\varphi_S$ are constant phases. Thus the basic STIRAP differs from other procedures that involve only two quantum states altogether or which involve only a single pulsed field or which employ fields with variable carrier frequencies (obtainable from time-varying phases $\varphi$).

In writing the field in this way one assumes that the field is classical, i.e. sufficiently strong that alteration by one photon has negligible effect on the field and the formalism of quantized

[4] Some authors refer to what is here written $\Omega/2$ as "Rabi frequency", often using the symbol $\Omega$. Some authors express "Rabi frequency" in Hz = $2\pi$ rad/sec.

[5] The magnitude of the electric-field amplitude $\mathcal{E}(t)$ in SI units obtains from the radiation intensity $I(t)$ as $|\mathcal{E}(t)| = \sqrt{2I(t)/(c\epsilon_0)}$, where $c$ is the vacuum speed of light and $\epsilon_0$ is the permittivity of free space.

electromagnetic fields [Dir27, Hei54, Lou73] is not needed. This is a satisfactory approximation for excitation by laser beams, but not for interaction with cavity fields, whose description requires quantized fields; see **Sec.** 1.3.12.

With electric-dipole interaction of eqn. (1.12) and the field of eqn. (1.13) the Rabi frequencies are evaluated from the formulas

$$\hbar\Omega_P(t) = -\mathbf{d}_{12}\cdot\mathbf{e}_P(t)\mathcal{E}_P(t), \qquad \hbar\Omega_S(t) = -\mathbf{d}_{23}\cdot\mathbf{e}_S(t)\mathcal{E}_S(t), \tag{1.14}$$

where $\mathbf{d}_{nm} = \langle\psi_n|\mathbf{d}|\psi_m\rangle$ is the electric-dipole transition moment between states $n$ and $m$. Numerical values for given pulse intensity $I(t)$ obtain from the formula [Sho90]

$$|\Omega| = 2.2068\times 10^9\,\mathrm{sec}^{-1}\,\sqrt{I[\mathrm{W\ cm}^{-2}]}\ \times\frac{|\mathbf{d}\cdot\mathbf{e}|}{ea_0}, \tag{1.15}$$

where $ea_0 = 2.54176$ debye is the atomic unit of dipole moment. That is, the Rabi frequencies depend explicitly on the relative orientations of the atom or molecule and the electric-field direction. For other multipole transitions (e.g. magnetic dipole or electric quadrupole) the Rabi frequency is proportional to the product of a multipole moment and a field amplitude. For an $n$-photon transition the Rabi frequency is proportional to a polarizability and the product of $n$ field amplitudes.

**Energy ranking.** It is important to note that by using the rotating-wave picture the original ranking of the quantum-state energies $E_n$ becomes immaterial: the basic equations (1.10)-(1.11) apply equally well to any ordering of undisturbed energies $E_n$, as does the STIRAP mechanism so long as the system starts in a state at the end of the chain, traditionally taken to be state 1 (see **Fig.** 1.2):

- a *ladder* ( or $\Xi$ or *cascade* ): $E_1 < E_2 < E_3$ ,
- a *lambda* (or $\Lambda$ or *folded*): $E_1, E_3 < E_2$,
- a *letter-V*: $E_1, E_3 > E_2$ .

In the classification of lambda and letter-V linkages the energy-ordering of states 1 and 3 does not matter: $E_1 < E_3$, $E_1 = E_3$ or $E_1 > E_3$ are all possible with the same $\mathsf{W}(t)$.

**Selectivity.** In writing eqn. (1.11) it is assumed that because of frequency selectivity and polarization-based selection rules [Sho90, Sho11] *each field acts only on a single transition*, as indicated in eqn. (1.4) and **Fig.** 1.1. (Breakdown of this rule is discussed in [Una00a].) The two transitions are characterized, in part, by the *single-photon detunings* $\Delta_P$ and $\Delta_S$, defined by their frequency mismatch with the appropriate Bohr transition frequency $\omega_{nm}$,

$$\Delta_P = \omega_{12} - \omega_P, \qquad \Delta_S = \omega_{23} - \omega_S, \qquad \hbar\omega_{nm} \equiv |E_n - E_m|. \tag{1.16}$$

These detunings are assumed to be much smaller than the carrier frequencies; they vanish for resonant transitions.

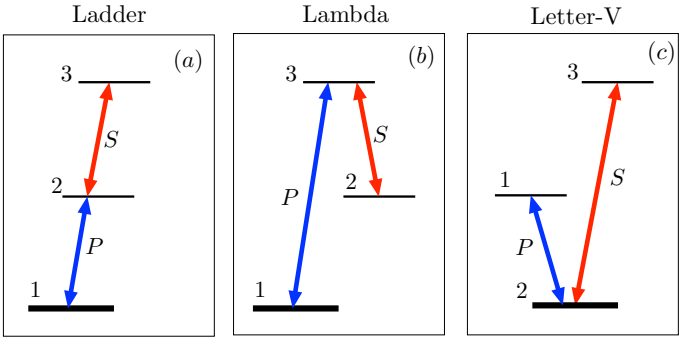


Fig. 1.2. The linkage patterns of $(a)$ Ladder, $(b)$ Lambda and $(c)$ Letter-V, showing $P$ and $S$ linkages. The relative ordering of energies $E_1$ and $E_3$ does not matter when using the RWA. In the letter-V linkage initial population is in the middle of the chain, state 2, whereas in the other patterns state 1 has the initial population.

**Detuning.** When used with pulsed electromagnetic fields the $\Delta$ of eqn. (1.11) is the (constant) single-photon detuning

$$\Delta = \Delta_P. \tag{1.17}$$

A characteristic of STIRAP, as defined here, is that $\Delta$ does not vary with time. Some other coherent-excitation schemes require time-varying detuning, typically a linear sweep, cf. **Sec.** 4.

**Two-photon resonance.** The condition for two-photon resonance, eqn. (1.3), needed for STIRAP and assumed for the $\mathsf{W}(t)$ of eqn. (1.11), implies that the $S$-field detuning for a given $P$ field detuning be

$$\Delta_S = \begin{cases} \Delta_P, & E_1, E_3 < E_2, \quad \textit{lambda linkage,} \\ -\Delta_P, & E_1, < E_2 < E_3, \quad \textit{ladder linkage.} \end{cases} \tag{1.18}$$

**Population loss.** As noted in **Sec.** 1.3.9, the real-valued $\Delta$ is often replaced by a complex number,

$$\Delta \to \Delta - \mathrm{i}\Gamma/2, \tag{1.19}$$

to model spontaneous emission from the excited state 2 and subsequent probability loss $-dP_2/dt$ at the rate $\Gamma P_2$. The population loss may also come from photoionization (or photodissociation) that takes the system into a continuum of kinetic energy states. In that situation $\Gamma$ may be time dependent. In all such cases the irreversible loss from a discrete state is into a continuum of states; the discrete excited state is "embedded" into a continuum, see **Sec.** 2.5. By setting $\Gamma = 0$ we are assuming that the times of interest are appreciably shorter than the radiative lifetime of the excited state.

**Degeneracy.** The spectroscopic applications of Raman scattering dealt with *degenerate* quantum states (i.e. states sharing a common energy but distinguishable by orientation) treated collectively as *energy levels* subject to *rate equations* for populations (see **App.** D and [Ack77]). Descriptions of coherent excitation, as in the present discussion, must treat each energy state separately; see **Sec.** 1.3.14.

### 1.2.2 Pulse ordering

**Labels.** **Section** 1.1 gives the original motive for the $P$ and $S$ terminology in the context of Raman processes. More generally the labeling of states 1 and 3 and of pulses $S$ and $P$ has the following meaning: by definition state 1 holds the initial population and state 3 is the *target state* for population transfer. The $P$ field is *primary*, in the sense of coupling the initially populated state to the excited state. The $S$ field is then *secondary*, meaning it has no connection to the initially populated state. In EIT the $P$ field is a *probe* altered by the *strong* $S$ field, see **Sec.** 5.1. In stimulated-emission pumping (SEP) the $S$ field is referred to as the *dump* field [Kit81,Ham86].

**Simultaneous pulses.** When simultaneous resonant pulses occur in a multi-state system (i.e. a linkage pattern with all detunings null, $\Delta_n = 0$), all with a common time variation $f(t)$, it is possible to introduce a scaled temporal variable $\tau(t)$ that incorporates all of the pulse shape, via the definition $d\tau = f(t)dt$. The dynamics is then described by a constant RWA Hamiltonian [Sho90, Sho11]. Under such conditions the time evolution exhibits Rabi oscillations rather than monotonic population transfer; see **Sec.** 2.9.2.

**Delayed pulses.** When the pulses are not simultaneous it is necessary to distinguish two orderings of the individual pulses. *Intuitive* pulse-ordering means that the primary field $P$ acts first, to move population from populated state 1 into excited state 2. *Counter-intuitive* ordering means that the secondary field $S$ precedes the primary field $P$; The earliest pulse, prior to the arrival of the $P$ pulse, has no direct effect on the initial population in state 1. The terminology of "intuitive" and "counter-intuitive" originated with understanding of incoherent radiative procedures that first move population into state 2 and then into state 3, [Sho95b]. The STIRAP process requires coherent radiation and relies on a counter-intuitive pulse sequence, with $S$ before $P$.

### 1.2.3 Two views of STIRAP

The RWA Hamiltonian matrix $\mathsf{W}(t)$, supplemented with initial conditions, provides only part of the definition of STIRAP. There are two complementary (in the Bohr sense) ways of viewing the remaining definition of STIRAP, related to the required pulse sequence.

**View 1.** One approach is a pragmatic, experimental viewpoint: there are two pulses, with carrier frequencies $\omega_P$ and $\omega_S$ adjusted for two-photon resonance, eqn. (1.18), to produce the diagonal elements of the matrix $\mathsf{W}(t)$ of eqn. (1.11). Population initially resides in state 1. The $S$ pulse, linking states 2-3, precedes and overlaps the $P$ pulse, which links states 1-2. The result of this pulse sequence will be complete population transfer $1 \rightarrow 3$ with never any appreciable excitation of state 2, if the variation of $\mathsf{W}(t)$ is sufficiently slow (i.e. the time evolution is *adiabatic*, cf. **App.** G). Figure 1.3 illustrates a simulation of this process.

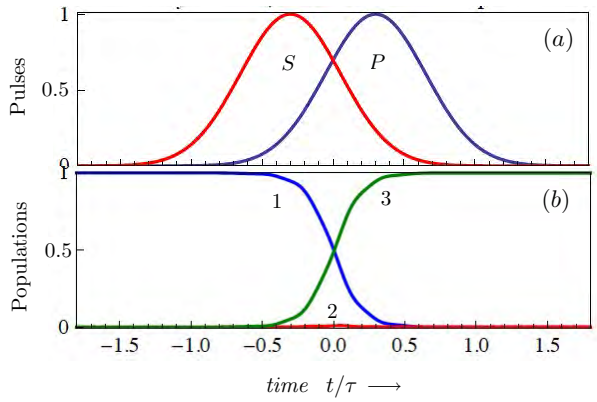


Fig. 1.3. Example of STIRAP dynamics for fully resonant Raman linkages, $\Delta = 0$. $(a)$ The pulsed Rabi frequencies $\Omega_S(t)$ and $\Omega_P(t)$, normalized to unit peak value. $(b)$ The populations $P_n(t)$. When the time evolution is adiabatic, as in this simulation, there is complete population transfer from state 1 to state 3 and negligible population in state 2, a signature of STIRAP. Times are in units of the pulse-width $\tau$. Theoretically, in the limit of perfect adiabatic evolution, state 2 would always remain unpopulated; but see **App.** G. Similar plots first appeared in [He90, Kuh92, Sho92b]; see also [Ber98, Vit01a, Vit01b, Sho08, Sho11].

**View 2.** The other approach is from a theoretical viewpoint, of Hilbert space and the three time-dependent eigenvectors[6] $\mathbf{\Phi}_\mu(t) \equiv |\mu\rangle$ of the instantaneous RWA Hamiltonian matrix $\mathsf{W}(t)$,

$$\mathsf{W}(t)\mathbf{\Phi}_\mu(t) = \varepsilon_\mu(t)\mathbf{\Phi}_\mu(t), \tag{1.20}$$

known variously as *adiabatic states* or *dressed states*, with *adiabatic eigenvalues* (also known as *quasi-energies*) $\varepsilon_\mu(t)$. These contrast with the original undisturbed *bare states* associated with basis vectors $\psi_n$ and $\psi'_n(t)$; their eigenvalues are the diagonal elements of $\mathsf{W}(t)$, known as *diabatic* eigenvalues. The adiabatic eigenvectors do not individually obey the TDSE, but they can be used to construct superpositions that do. The STIRAP process relies on a particular eigenvector of the $\mathsf{W}(t)$ of eqn. (1.11) (the population-trapping dark state, cf. **Secs.** 1.3.9-1.3.10 and 2.3):

$$\mathbf{\Phi}_0(t) = \frac{1}{\Omega_{\rm rms}(t)} \begin{bmatrix} \Omega_S(t) \\ 0 \\ -\Omega_P(t) \end{bmatrix}, \qquad \Omega_{\rm rms}(t) = \sqrt{\Omega_P(t)^2 + \Omega_S(t)^2}, \tag{1.21}$$

which, for the RWA Hamiltonian of eqn. (1.11), has a constant null eigenvalue, $\varepsilon_0(t) = 0$ (but see **App.** E.1). To produce the STIRAP process we adjust the Rabi frequencies such that initially,

[6] The boldface greek-letter notation $\mathbf{\Phi}_\mu$ emphasizes the mathematical similarity with Hilbert-space vectors $\mathbf{\Psi}$ and $\boldsymbol{\psi}_n$. The Dirac notation $|\mu\rangle$ is better suited to labels on curves, where the ket notation $|\cdots\rangle$ makes clear the meaning of plus and minus signs.

at time $t = 0$, the statevector $\mathbf{\Psi}(0)$ coincides with $\mathbf{\Phi}_0(0)$ and with the initially populated bare state 1,

$$\mathbf{\Psi}(0) \approx \mathbf{\Phi}_0(0) \approx \psi_1'(0), \qquad \text{for } |\Omega_S(0)| \gg |\Omega_P(0)|. \tag{1.22}$$

Then we control changes to the two Rabi frequencies such that, at the conclusion of the pulse sequence, $\mathbf{\Phi}_0(T)$ lies along the 3 axis, coinciding (apart from phase) with bare state 3:

$$\mathbf{\Psi}(T) \approx \mathbf{\Phi}_0(T) \approx -\psi_3'(T), \qquad \text{for } |\Omega_P(T)| \gg |\Omega_S(T)|. \tag{1.23}$$

We make the changes sufficiently slowly (adiabatically) that the statevector $\mathbf{\Psi}(t)$ remains always aligned with $\mathbf{\Phi}_0(t)$; see **App.** G. The result of this controlled Hilbert-space rotation of the adiabatic eigenvector is complete population transfer, $1 \to 3$. Because $\mathbf{\Phi}_0(t)$ has no component of state 2, there will never be appreciable population in this intermediate state.

The variation in the Hilbert-space structure of $\mathbf{\Phi}_0(t)$ during the course of a STIRAP process is most readily tracked by means of the *mixing angle* $\theta(t)$, defined by the ratio of Rabi frequencies[7]:

$$\mathbf{\Phi}_0(t) = \begin{bmatrix} \cos\theta(t) \\ 0 \\ -\sin\theta(t) \end{bmatrix}, \qquad \sin\theta(t) = \frac{\Omega_P(t)}{\Omega_{\rm rms}(t)}, \qquad \cos\theta(t) = \frac{\Omega_S(t)}{\Omega_{\rm rms}(t)}. \tag{1.24}$$

By controlling the $P$ and $S$ fields the experimenter rotates the mixing angle from 0 to $\pi/2$, thereby shifting the dominant component of $\mathbf{\Phi}_0(t)$ from state 1 to state 3. **Figure** 1.4 shows the Hilbert-space motion of the vectors $\mathbf{\Phi}_\mu(t)$ associated with STIRAP. When the statevector $\mathbf{\Psi}(t)$ remains aligned with $\mathbf{\Phi}_0(t)$, following its Hilbert space motion [an example of *adiabatic following* (AF); see **Sec.** 2.2.3] then the populations transfer from state 1 to state 3.

The adiabatic eigenvectors are used extensively in discussions of STIRAP, but other approaches have uses for depicting the dynamics. **Sections** 1.3.5, 1.3.9 and 2.2.8 define other useful rotating-coordinate systems for viewing three-state dynamics. [Keep in mind that the bare-state basis vectors of eqn. (1.7) already incorporate rotations at carrier frequencies.]

### 1.2.4 Representative STIRAP results

Simulations of examples of the TDSE, for very general pulse shapes (and large numbers of quantum states) are readily obtained using commercial software such as Mathematica ® (my own choice) or Matlab ® , with any personal computer platform. Numerous articles have displayed examples of the several variables associated with STIRAP processes, showing successful population transfer, as in **Fig.** 1.3. Mostly these illustrations have been for situations in which the two pulses, though offset by a delay, have the same time dependence (typically Gaussian) and the same Rabi-frequency peak value. Such restrictions are not necessary. **Figure** 1.5 shows an example of simulation for Rabi-frequency pulses of finite duration that have unequal peak values and negative single-photon detuning, $\Delta = -5$. The top frame, $(a)$, shows the relative values of the two Rabi frequencies. Frame $(b)$ shows the three adiabatic eigenvalues, labeled $|-\rangle, |0\rangle, |+\rangle$. The central curve (dashed) is the constant associated with $|0\rangle$. Frame $(c)$ shows the mixing angle

[7] The more concise definition $\theta = \arctan \Omega_P/\Omega_S$ must be used with caution when making numerical evaluations.

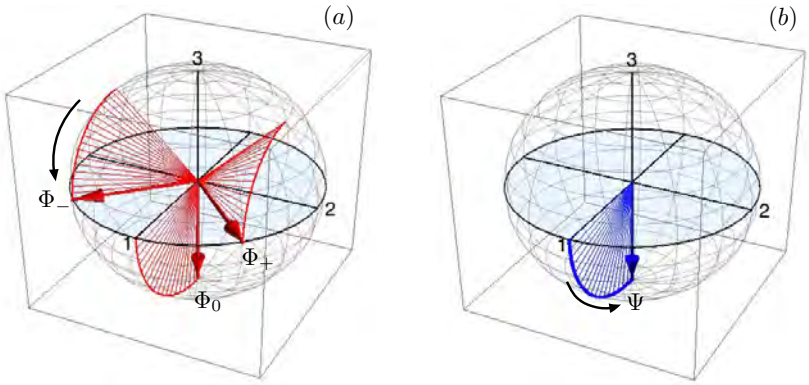


Fig. 1.4. Examples of Hilbert-space rotations into final locations, associated with STIRAP. $(a)$ The three adiabatic eigenvectors rotate as shown: The eigenvectors $\mathbf{\Phi}_+$ and $\mathbf{\Phi}_-$ rotate onto the equatorial plane, the eigenvector $\mathbf{\Phi}_0$ rotates in the 1,3 plane from the 1 axis to the -3 axis. $(b)$ The statevector $\mathbf{\Psi}$, maintaining alignment with $\mathbf{\Phi}_0$, rotates from the 1 axis to the -3 axis.

$\theta(t)$, varying monotonically from 0 to $\pi/2$. The bottom frame, $(d)$, shows the populations $P_n(t)$. With the chosen parameters there is some small temporary population in state 2, but overall there is complete population transfer, as in **Fig.** 1.3.

**The mixing angle.** The pulses used for the simulation of **Fig.** 1.5 have finite duration (see **App.** H). The change of mixing angle, and population, takes place only during a finite interval when both pulses are present. The grayed-hashed portions of the frames mark times when only a single field is present. These intervals, and those in which one field is very weak, are irrelevant for the population dynamics.

**The system point.** The conditions for the simulations shown in **Fig.** 1.5 are such that the time evolution is adiabatic and the statevector remains closely aligned with (follows) the vector $\mathbf{\Phi}_0(t)$. The dynamics can be traced by a system point that traces its eigenvalue, $\varepsilon_0(t)$, in the plot of frame $(b)$. Initially and finally two of the eigenvalues are zero, those labeled $|0\rangle$ and $|+\rangle$; the third, $|-\rangle$ is equal to the detuning $\Delta$, here a negative number. For the choice of parameters of this calculation the time evolution is satisfactorily adiabatic, and the system point follows the null-eigenvalue adiabatic state, as indicated by the arrows.

## 1.3 STIRAP characteristics

A number of conditions are necessary for successful population transfer with the STIRAP process. Conversely, a system (quantum or classical) that undergoes STIRAP will exhibits several identifying characteristics. Following are some conditions that were considered very early, although subsequent work has, in some cases, altered the early restrictions. The reviews [Ber98, Vit01a, Vit01b] discuss the sensitivity of STIRAP to experimental conditions.

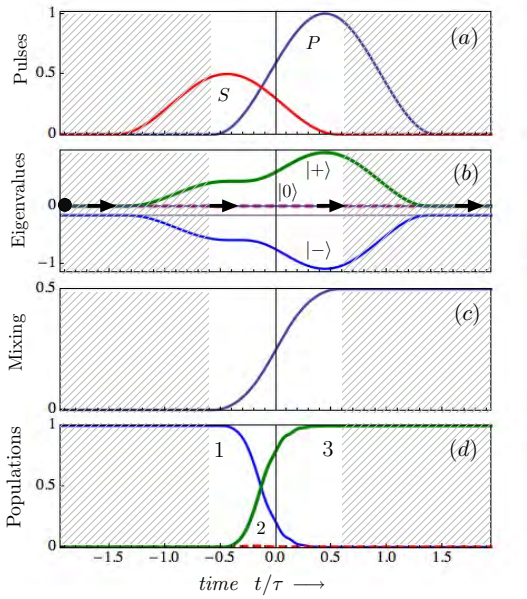


Fig. 1.5. Example of STIRAP with unequal finite-duration pulses and static nonzero single-photon detuning $\Delta$. $(a)$ Relative Rabi frequencies, showing $S$ before $P$. $(b)$ Relative adiabatic eigenvalues $\varepsilon_\mu(t)$. The arrows show the adiabatic motion of the system point, positioned initially at the large dot on the curve $|0\rangle$. $(c)$ Mixing angle $\theta(t)$ in units of $\pi$. The pulse sequence produces a $\theta(t)$ change from 0 to $\pi/2$. $(d)$ Populations $P_n(t)$. The grayed portions of the frames mark intervals when only a single field is present; they have no effect on the population change.

### 1.3.1 Coherence

The dynamics described here for the STIRAP process for a quantum system is *coherent*, involving phase-preserving changes described by the time-dependent Schrödinger equation (1.9) and a statevector $\boldsymbol{\Psi}(t)$ [Sho90, Sho11]. Here "coherent" means that both the quantum system (atom or molecule) and the excitation field (detuning, Rabi frequencies and phases $\varphi$) are free from random uncontrollable environmental disturbances. Light from thermal sources, even when spectrally filtered, does not have the required coherence. It is possible to treat some irregular stochastic influences, including rigorous treatment of spontaneous emission, by means of a *density* (or *statistical*) *matrix* $\boldsymbol{\rho}(t)$ formed from bilinear products of probability amplitudes[8],

$$\rho_{nm}(t) = C_n(t)C_m(t)^*, \tag{1.25}$$

but these effects cannot be large without destroying coherence and preventing the adiabatic following that is a key element of STIRAP. For a discussion of theoretical and experimental consequences of laser bandwidth on STIRAP see [Kuh92].

[8]Many authors incorporate the phases $\zeta_n(t)$ into the definition of $\boldsymbol{\rho}(t)$.

A signature of coherent excitation by *steady* (unpulsed) fields of constant frequency is the occurrence of Rabi oscillations in populations [All75, Kni80, Sho11], not present with incoherent excitation and rate-equation descriptions of change (see **App.** D).

A combination of steady-field coherent excitation along with (or followed by) incoherent spontaneous emission occurs in the technique of *optical pumping* [Hap72, Kit81, He90, Sho11]. The final properties of such a process are generally not a single quantum state; a density matrix must be used to treat such processes.

### 1.3.2 Three states and two pulsed fields; The RWA

Historically, the basic STIRAP process involved three quantum states and two pulsed classical radiation fields, with the statevector adiabatically following a single (dark) adiabatic state (see **Sec.** 1.3.9). **Section** 6.4 mentions extensions to more states and interactions.

The two fields may be distinguished either by wavelength (as in the original Raman process) or by polarization, as occurs in a Raman transition between Zeeman sublevels of a degenerate ground level, see **Fig.** 1.10$(a)$ in **Sec.** 1.3.14. The pulsed electromagnetic fields typically originate in one of two ways:

- The pulse shape may originate with the variation of field intensity as beam particles travel across a directed stationary laser beam, as was the case with the first STIRAP experiments [Gau90, Kuh92, Mar96]. Such pulses have been modeled as Gaussians. Particles traveling with different velocities will experience faster or slower time variation.

- Alternatively the field envelopes may come from crafted laser-pulses acting on stationary quantum systems, as was done with nanosecond pulses by [Sch93, Hal96]. On such a time scale the particles in a vapor or liquid can be regarded as stationary. With microwave pulses it is feasible to program the exact electric field of tens of cycles, thereby creating a discrete approximation of quite general amplitude, phase and frequency variation — an example of arbitrary waveform generation (AWG). The crafted femtosecond laser pulses created by frequency-component manipulation [Fro83, Wei90, Bri03, Bri04] are generally of too short duration for use as STIRAP pulses (but see the work by Shapiro on femtosecond pulse trains [Sha07]).

**The RWA.** When dealing with radiation fields for use with STIRAP it is important to use the RWA when setting up the mathematical description. This approximation assumes that any statevector changes, caused by Rabi frequencies and detunings, are much slower than the field carrier frequencies, justifying the replacement (see [All75, Sho90, Sho11])

$$1 + \exp[\pm \mathrm{i}2\omega t] \to 1, \tag{1.26}$$

where $\omega$ is either of the carrier frequencies. This approximation is not required when the interactions responsible for the Rabi frequencies are not those of electromagnetic transitions (e.g. electric or magnetic dipole transitions or multiphoton transitions) but are slowly varying quasistatic interactions, as noted in **Sec.** 6.8. When the Rabi frequency or the detuning becomes comparable to the Bohr frequency it is necessary to include counter-rotating terms in the Hamiltonian.

**Intensity constraints.** To accomplish coherent excitation the fields (and corresponding Rabi frequencies) cannot be too weak, because their action must be completed before inevitable phase-disrupting influences affect the system, but they cannot be too strong (as are commercial lasers used for welding and cutting), or else they will rapidly induce nonlinear processes that will ionize or dissociate the system of interest and destroy the initial quantum states. Electric fields must therefore be much less than the electric field that holds electrons within an atom or molecule.

**Other systems.** As noted in **Sec.** 6.8, there are many physical systems, both quantum mechanical and classical, that deal with ODEs like eqns. (1.10) and (1.11) for three variables and two pulsed interactions. In these systems there can be found analogs of all of the properties associated with traditional STIRAP.

### 1.3.3 Double (two-photon) resonance

Although it is not necessary for STIRAP that the individual-field carrier frequencies $\omega_P$ and $\omega_S$ be resonant with their respective Bohr frequencies $\omega_{12}$ and $\omega_{23}$, they must maintain the two-photon resonance condition (1.18). Thus STIRAP is an extension of work on optical double resonance [Bro52, Whi76], but with pulsed fields that are not simultaneous.

From a theoretical viewpoint, the critical property of two-photon resonance is the presence of the two null elements on the diagonal of the $3 \times 3$ matrix $\mathsf{W}(t)$ for the basic ODEs of STIRAP, eqn. (1.10). As noted in **Sec.** 6.8 any three ODEs of this form have the potential for exhibiting the efficient adiabatic change of system variables associated with STIRAP.

### 1.3.4 Two-photon detuning

Although STIRAP, as defined here, requires the statevector to maintain continuously alignment with the dark adiabatic state, and hence requires two-photon resonance, it is quite possible to achieve excellent population transfer when this resonance condition is violated, so that the RWA Hamiltonian, with allowance for loss from states 2 and 3, is

$$\mathsf{W}(t) = \frac{1}{2} \begin{bmatrix} 0 & \Omega_P(t) & 0 \\ \Omega_P(t) & 2\Delta_2 - \mathrm{i}\Gamma_2 & \Omega_S(t) \\ 0 & \Omega_S(t) & 2\Delta_3 - \mathrm{i}\Gamma_3 \end{bmatrix}, \tag{1.27}$$

where, for radiative interactions, the *cumulative detunings* are

$$\Delta_2 = \Delta_P \tag{1.28}$$

$$\Delta_3 = \begin{cases} \Delta_P - \Delta_S, & E_1, E_3 < E_2, \quad \textit{lambda linkage,} \\ \Delta_P + \Delta_S, & E_1, < E_2 < E_3, \quad \textit{ladder linkage}. \end{cases} \tag{1.29}$$

The adiabatic curves now may have intersections ("avoided crossings") at which times the statevector shifts alignments between degenerate adiabatic states, rather than maintaining alignment with a single adiabatic state [Dan94, Rom97, Gri01, Yat02a]. The process is not STIRAP, but the population histories may be hardly distinguishable from those of STIRAP. **Figure** 1.6 shows an example, with simulations that use larger pulse areas than are used in experiments, in order to achieve good adiabatic evolution.

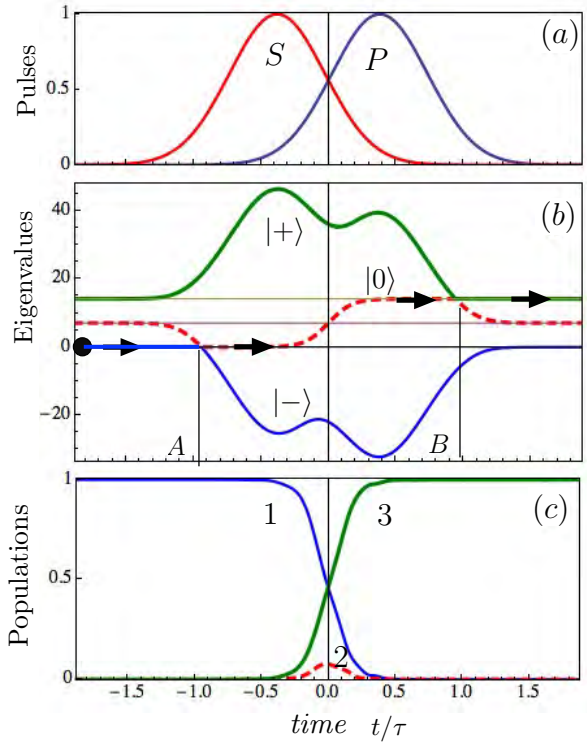


Fig. 1.6. Simulation of population transfer with two-photon detuning. $(a)$ Relative values of the two Rabi frequencies, $S$ before $P$. $(b)$ The adiabatic eigenvalues for states $|-\rangle$, $|0\rangle$ (dashed) and $|+\rangle$. Arrows show the course of the system point, which follows an adiabatic state except at two curve intersections, $A$ and $B$, when the point follows a diabatic curve. $(c)$ The population histories $P_n(t)$. Population is transferred from state 1 to state 3, with brief non-adiabatic presence of state 2. These simulations use identical Gaussian pulses, of areas $20\pi$, and detunings $\Delta_2 = 0.1\Omega_0, \Delta_3 = 0.2\Omega_0$.

### 1.3.5 STIRAP and torque

A statevector undergoing coherent change maintains unit length, and hence its motion in Hilbert space is a generalized rotation. When the excitation is fully resonant, meaning $\Delta_P = \Delta_S = 0$, the TDSE (1.10) can be presented in the form of a three-dimensional torque equation,

$$\frac{d}{dt}\mathbf{r} = \boldsymbol{\Upsilon}\times\mathbf{r}, \tag{1.30}$$

in which an *angular-velocity vector* $\boldsymbol{\Upsilon}$ (also called the *torque vector*) acts to rotate the vector $\mathbf{r}$. For that purpose consider the TDSE for three states coupled by three resonant interactions,

$$\mathrm{i}\frac{d}{dt}\begin{bmatrix} C_1 \\ C_2 \\ C_3 \end{bmatrix} = \tfrac{1}{2}\begin{bmatrix} 0 & \Omega_P & \mathrm{i}\Omega_Q \\ \Omega_P & 0 & \Omega_S \\ -\mathrm{i}\Omega_Q & \Omega_S & 0 \end{bmatrix}\begin{bmatrix} C_1 \\ C_2 \\ C_3 \end{bmatrix}, \tag{1.31}$$

To rewrite this as a torque equation we introduce new independent variables differing from $C_n$ by a phase, $[r_1, r_2, r_3]^T = [C_1, \mathrm{i}C_2, -C_3]^T$, so that the TDSE reads

$$\frac{d}{dt}\begin{bmatrix} r_1 \\ r_2 \\ r_3 \end{bmatrix} = \tfrac{1}{2}\begin{bmatrix} 0 & -\Omega_P & \Omega_Q \\ \Omega_P & 0 & -\Omega_S \\ -\Omega_Q & \Omega_S & 0 \end{bmatrix}\begin{bmatrix} r_1 \\ r_2 \\ r_3 \end{bmatrix}. \tag{1.32}$$

This is an example of a torque equation in which the components of the angular-velocity vector $\boldsymbol{\Upsilon}$ are half the Rabi frequencies,

$$\boldsymbol{\Upsilon} = \tfrac{1}{2}[\Omega_S, \Omega_Q, \Omega_P]^T. \tag{1.33}$$

**Adiabatic following.** The vector $\mathbf{r}$, of unit length, will remain aligned with (adiabatically follow) the angular-velocity vector if initially the two vectors are either parallel or antiparallel,

$$\boldsymbol{\Upsilon} \cdot \mathbf{r} = \pm|\boldsymbol{\Upsilon}|\mathbf{r}, \quad \text{so that } \boldsymbol{\Upsilon} \times \mathbf{r} = 0, \tag{1.34}$$

and the subsequent changes of $\boldsymbol{\Upsilon}$ are sufficiently slow (adiabatic); see **App.** G. That is, $\mathbf{r}$ is an instantaneous right eigenvector of the angular-velocity vector[9].

**Examples.** The required adiabatic vectors are simplest to obtain when, as in STIRAP, there are only two interactions. For example, let $\Omega_Q = 0$. Then the required adiabatic-following vector $\mathbf{r}$ is obtained from the following expression for $\boldsymbol{\Upsilon} \times \mathbf{r} = 0$ with $\mathbf{r} = \boldsymbol{\Upsilon}/|\boldsymbol{\Upsilon}|$:

$$\begin{bmatrix} 0 & -\Omega_P & 0 \\ \Omega_P & 0 & -\Omega_S \\ 0 & \Omega_S & 0 \end{bmatrix}\begin{bmatrix} \Omega_S \\ 0 \\ \Omega_P \end{bmatrix} = 0. \tag{1.35}$$

This is the torque equation appropriate to STIRAP, and it yields the conventional (dark) adiabatic eigenvector that has null component $r_2 = \mathrm{i}C_2$. Adiabatic change of the angular-velocity vector from strong $S$ field, $\boldsymbol{\Upsilon} = [1, 0, 0]^T$ to strong $P$ field, $\boldsymbol{\Upsilon} = [0, 0, 1]^T$, will take the vector $\mathbf{r}$ from $[1, 0, 0]^T$ to $[0, 0, 1]^T$, or $C_3 = -1$. This is STIRAP, viewed as a solution to the torque equation. However, any change in mixing angle necessarily introduces some nonzero component of $r_2 = \mathrm{i}C_2$; see **App.** G.

Alternatively, let $\Omega_S = 0$. Then the relevant equation for adiabatic following is

$$\begin{bmatrix} 0 & -\Omega_P & \Omega_Q \\ \Omega_P & 0 & 0 \\ -\Omega_Q & 0 & 0 \end{bmatrix}\begin{bmatrix} 0 \\ \Omega_Q \\ \Omega_P \end{bmatrix} = 0. \tag{1.36}$$

Now the vector used for adiabatic following has only components of states 1 and 2, between which adiabatic transfer will occur as the two fields shift from large $\Omega_Q$ to large $\Omega_P$. This too is STIRAP, but with a relabeling of the states and the interactions.

[9] Although $\boldsymbol{\Upsilon}$ takes its elements from the RWA Hamiltonian W, the construction of $\mathbf{r}$ as an eigenvector of $\boldsymbol{\Upsilon}$ is not the same thing as constructing an eigenvector of W.

### 1.3.6 Adiabatic following

Whereas general schemes of coherent excitation depend upon details of pulse shapes, most notably the *temporal pulse area*[10]

$$\mathcal{A} = \int_{-\infty}^{+\infty} dt\, \Omega(t), \tag{1.37}$$

adiabatic processes are generally insensitive to such details — they are termed *robust*. The success of a STIRAP process depends primarily upon the success of maintaining alignment of the statevector $\mathbf{\Psi}(t)$ with the adiabatic eigenvector $\mathbf{\Phi}_0(t)$ (or the vector $\mathbf{r}$ with the angular-velocity vector $\mathbf{\Upsilon}$) during all changes to the RWA Hamiltonian $\mathsf{W}(t)$. The changes must therefore be slow, in some sense: they must be adiabatic [Mes62, Mor64, Loy74, Gau90]. **Appendix** G gives details for the STIRAP process.

Such statevector changes are examples of *adiabatic following* and, when they produce population transfer, they are examples of *adiabatic passage* (AP)[11] between quantum states, cf. **Sec.** 2.2.5. The original research that used the STIRAP mechanism, e.g. [Gau88, Kuk89, Gau90, Ber98], focused on complete population transfer, i.e. adiabatic *passage*.

Subsequent work, using much of the formalism developed for STIRAP, demonstrated creation of coherent superpositions of quantum states, i.e. adiabatic *following*, e.g. [Vit99] and **Sec.** 6.1, but there has been no suggestion to use the acronym STIRAF for this.

Note: Originally the term "following" referred to continuing alignment of a macroscopic dipole moment with an effective field, or of the Bloch vector with an angular-velocity vector whose components came from a Hamiltonian, cf. **Secs.** 2.2.2 and 2.2.3. In the context of STIRAP the term "following" usually refers to continual alignment of the statevector with an adiabatic eigenvector or eigenstate, but equally well it can refer to alignment with the angular-velocity vector of a torque equation, see **Sec.** 1.3.5.

### 1.3.7 Adiabatic condition

**Appendix** G discusses the conditions on the RWA Hamiltonian that are needed for adiabatic following. Manifestations of non-adiabatic evolution of the three state system are evident in two ways: the failure of $P_3(t)$ to be unity at the termination time $T$ of the pulse sequence and the failure of $P_2(t)$ to be zero at all times. Let us define these errors as

$$\epsilon_3 = 1 - P_3(T), \qquad \epsilon_2 = \text{Max } P_2(t). \tag{1.38}$$

As the pulse area becomes larger, each of these errors diminishes. By choosing some error criteria an experimenter is led to a requirement on minimum pulse area $\mathcal{A}$. Reviews [Ber98, Vit01b] have suggested an area of at least 10 (or $3\pi$) as suitable; see **App.** G. **Figure** 1.7 shows an example of the connection between error and pulse area for two identically shaped Gaussian pulses, suitably

[10] In a two-state system resonantly driven by a monochromatic field complete population transfer will occur when $\mathcal{A}$ is an odd-integer multiple of $\pi$, a so-called *pi pulse*.

[11] It has been suggested to me that although "passage" originally meant the complete transfer of population the term could equally well describe the creation of *any* predetermined final state, of which a pure state is but one example. Consistent with that notion the creation of a superposition state could be called "adiabatic passage" in addition to being a halted "adiabatic following".

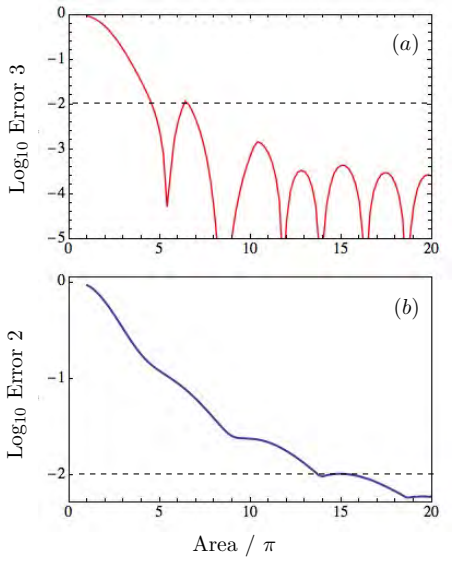


Fig. 1.7. $(a)$ The error $\epsilon_3$ $(b)$ The error $\epsilon_2$, both as a function of temporal pulse area for two near-optimally overlapping Gaussian pulses. Dashed horizontal lines mark errors of $10^{-2}$.

offset in time, and a lossless RWA Hamiltonian that has both single-photon and two-photon resonance: $\Delta_2 = \Delta_3 = 0$. The figure shows that, based on the requirement of an error less than $10^{-2}$ in population transfer, a pulse area larger than $4\pi$ is needed. To maintain a state-2 population less than $10^{-2}$ it is necessary to have a pulse area less than around $15\pi$. Somewhat different values would be obtained if the error $\epsilon_2$ is defined as the integrated population in state 2. The early use of STIRAP for preparation of preselected excited states found population transfer $P_3(T) = 0.9$ satisfactory, but applications in quantum-information processing require values differing from 1.0 by $10^{-5}$.

**Allowable detuning.** The single-photon detuning $\Delta_2$ does not enter the definition of the dark state and so it does not directly affect the possibility of complete population transfer as long as there is two-photon resonance, $\Delta_3 = 0$ . However, the detuning does affect the adiabaticity of the dynamics [Vit01a], adding coupling between $|+\rangle$ and $|-\rangle$ and thereby requiring a larger pulse area (and larger pulse energy) to maintain the same small error; see **App.** G. STIRAP therefore operates most efficiently on single-photon resonance, when $\Delta_2 = 0$ (and, of course, $\Delta_3 = 0$).

**Irreversible.** It is sometimes suggested (cf. **Sec.** 2.2.6 and [Pet82,Pet85]) that adiabatic changes should be reversible and that for any excitation to persist after passage of a pulse it is necessary that adiabatic evolution must be violated at some time — either the slow pulse buildup must be followed by a rapid cutoff or else there must by decoherence-inducing processes (e.g. collisions or photoionization) that negate the description of coherent excitation by the TDSE. This

expectation of reversibility attends excitation by a single pulse, or simultaneous pulses, but not the sequential overlapping pulses of STIRAP, for which the initial and final Hamiltonians differ: the STIRAP procedure demonstrably does *not* inevitably return all population to the initial state. Reversibility does occur if the $S-P$ sequence is followed by the reversed sequence $P-S$. This will return population from state 3 to state 1 and restore the initial conditions.

### 1.3.8 Pulse S before P; Counter-intuitive ordering

The counter-intuitive pulse timing, $S$ before $P$, is an essential aspect of STIRAP not present in the traditional double-resonance work. Because changes are adiabatic the shapes of the two envelopes are not important, within limits. The dynamics are most simply understood when the pulse envelopes have finite duration: the $P$ pulse-envelope is zero initially, when the $S$ pulse is nonzero, and the $S$ pulse-envelope is zero finally, when the $P$ pulse is nonzero. As the temporal pulse areas increase (and so adiabatic following is better satisfied) there is little effect on the population transfer produced by a pulse sequence having (negative) delay of around half the pulse width. However, there is a very notable variation with pulse area in the results for positive detuning, $P$ before $S$: for that regime Rabi oscillations are evident. **Figure** 1.8 shows, for a lossless and fully resonant system, $\Delta_2 = \Delta_3 = 0$, a set of plots of population transfer probability for four pulse areas, ranging from $5\pi$ to $20\pi$. As can be seen, when the pulse areas are an odd-integer multiple $\pi$ there occurs complete transfer for any large positive delay, whereas when the pulse areas are an even-integer multiple $\pi$ there is complete population return for any large delay.

To observe such variation of population transfer it is, of course, necessary that all the atoms be subject to the same temporal pulse area: they must all have the same dipole transition-moment and experience the same field. In experiments on molecular beams passing through laser beams, as in [Gau90], this condition does not hold: the molecules have a range of speeds and interact with a range of peak Rabi-frequencies, from the different rotational orientations (i.e. different magnetic quantum numbers) thereby blurring the pulse-area dependence. Furthermore there can be no loss from state 2: this would damp the Rabi oscillations.

**Transfer with $P$ before $S$.** The regime at negative delay is where adiabatic transfer (STIRAP) occurs. For zero delay there are Rabi oscillations that regularly place half the population into state 2 (see **Sec.** 2.9.2), and the final populations depend upon the temporal pulse areas. When there is large nonzero single-photon detuning, but two-photon resonance $\Delta_3 = 0$, it is possible to achieve complete population transfer with an intuitive ordering of pulses, independent of pulse area, but this is not STIRAP; see **Sec.** 6.2.

### 1.3.9 The dark state

In the traditional Raman or stimulated Raman processes the middle state of the three-state chain is an excited state, and thus it can undergo spontaneous emission, visible as fluorescence. Often such an emission will lead to a state other than states 1 or 3: the system will no longer be describable by a statevector in a closed three-dimensional Hilbert space[12] . It is customary to

[12] Allowance for spontaneous transitions amongst three states cannot be treated as a coherent process with a statevector. It requires a density matrix.

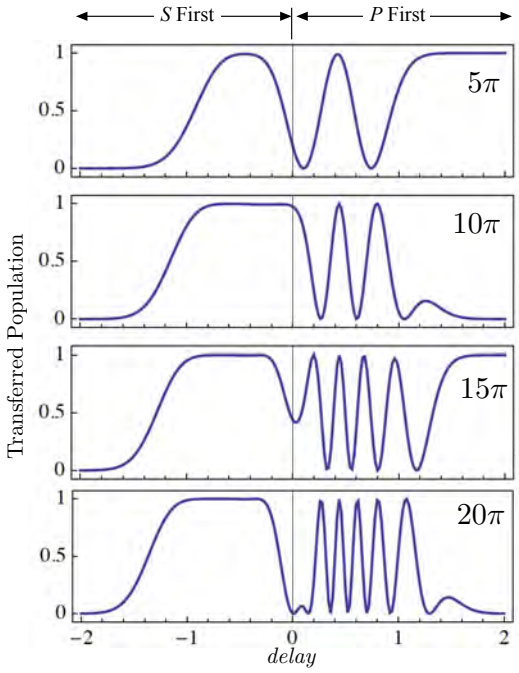


Fig. 1.8. Population transfer versus delay (in units of pulse width $\tau$) for lossless, fully resonant excitation, $\Delta_P = \Delta_S = 0$ and four choices of pulse areas: top to bottom $5\pi, 10\pi, 15\pi, 20\pi$. With $S$ first the population transfer is insensitive to pulse area, whereas with $P$ first the transfer depends on pulse area via Rabi oscillations. (When state 2 has loss, these oscillations are damped; for pulses longer than the lifetime they will therefore be absent.) The labels "$S$ first" and "$P$ first" assume population starts in state 1. The plots also apply for transfer from state 3 to state 1, with interchange of $S$ and $P$.

treat such probability loss by making the energy $E_2$ complex; the detuning $\Delta$ of eqn. (1.11) then becomes $\Delta - \mathrm{i}\Gamma/2$, and any population arriving in state 2 will produce a fluorescence signal.

Because the eigenvector $\mathbf{\Phi}_0(t)$ is always a coherent superposition of states 1 and 3 it never has any component of lossy state 2, and its components never produces fluorescence: it is a *dark state* [Ari96]. That is, the STIRAP process relies on a *time-varying dark state* to produce population transfer.

With the traditional (but arbitrary) choice of phases and energy zeros, eqn. (C.4a), the dark state has zero eigenvalue, so that $\mathsf{W}(t)\mathbf{\Phi}_0(t) = 0$, but that choice is by no means necessary; cf. **Fig.** 3.2 in **Sec.** 3.3 and **App.** E.1.

**The bright state.** The description of Hilbert-space statevector motion of a three-state system, such as that of the basic stimulated Raman Hamiltonian $\mathsf{W}(t)$ of eqn. (1.11), requires three orthogonal unit vectors. The simplest choice is the set of stationary vectors $\psi_n$ or the rotating vectors $\psi'_n(t)$ associated with physical states. The choice used for **Fig.** 1.4$(a)$ is the set of

instantaneous eigenvectors $\mathbf{\Phi}_\mu(t)$ of $\mathsf{W}(t)$. A third possibility retains the excited-state $\psi'_2(t)$ and the dark adiabatic state $\mathbf{\Phi}_0(t)$ as two of the coordinates. The resulting dark-bright set [Vit97c, Vit97d, Fle96, Fle99, Pas99, Kis02], with elements labeled dark state $(d)$, bright state $(b)$ and excited state $(e)$, are

$$\mathbf{\Phi}_d(t) = \cos\theta(t)\,\psi'_1(t) - \sin\theta(t)\,\psi'_3(t) \equiv \mathbf{\Phi}_0(t), \tag{1.39a}$$

$$\mathbf{\Phi}_b(t) = \sin\theta(t)\,\psi'_1(t) + \cos\theta(t)\,\psi'_3(t), \tag{1.39b}$$

$$\mathbf{\Phi}_e(t) = \psi'_2(t). \tag{1.39c}$$

The bright and dark unit vectors rotate in the 1,3 plane of Hilbert space, while the third unit vector remains fixed, along the 2 axis [trecall that the coordinate vectors $\psi'_n(t)$ are rotating with phases $\zeta_n(t)$].

**Two-state behavior.** When the mixing angle $\theta(t)$ remains constant, as it will when the $P$ and $S$ pulses have the same time dependence, the amplitudes in this basis obey the equations

$$\mathrm{i}\frac{d}{dt}C_b(t) = \tfrac{1}{2}\Omega_{\mathrm{rms}}C_e(t), \tag{1.40a}$$

$$\mathrm{i}\frac{d}{dt}C_e(t) = \tfrac{1}{2}\Omega_{\mathrm{rms}}C_b(t) + \Delta C_e(t), \tag{1.40b}$$

$$\mathrm{i}\frac{d}{dt}C_d(t) = 0, \tag{1.40c}$$

and the dynamics appears as that of a two-state system: the bright state embodies all of the interaction with the excited state. Section 2.10.1 discusses a second type of two-state behavior, occurring when the detuning $|\Delta|$ is very large.

### 1.3.10 Population trapping

The population that starts in state 1 and remains associated with the adiabatic state $\mathbf{\Phi}_0(t)$ does not undergo loss by spontaneous emission or by any other mechanism that is associated with excited state 2. This population remains *trapped*, locked in a *population trapping* state [Rad82]. **Appendix** E.2 discusses a very simple example of population trapping, in which the trapped state occurs because of a choice of coordinate system: a rotation of coordinates alters the magnetic-sublevel composition of an angular momentum state.

An important characteristic of the STIRAP process is that state 2 is not populated. One has a choice of making this either a condition or a consequence. If you require that the $S$ pulse precedes the $P$ pulse, and the detuning is constant, the particular adiabatic state of interest turns out to be the dark state. Alternatively, if you make the dark state, with its null population in state 2, the condition, then the counter-intuitive pulse sequence is a consequence along with constant detuning. In the sense of Euclid, axiomatizing geometry, you have a choice of your basic non-redundant axioms.

### 1.3.11 Coherence-free subspace

As long as the statevector remains well aligned with the dark state its motion is in a two-dimensional subspace of the three-dimensional Hilbert space, and its behavior can be described

by two-state variables. This dimensional reduction is a special case of more general dark states that contain no excited-state component, cf. **Sec.** E.3.

Even a well-isolated single $N$-state quantum system, such as an atom or molecule, inevitably interacts with an external environment over which an experimenter has no complete control. Random fluctuations of the environment act to alter the phases and amplitudes associated with the single system, in an uncontrollable way, a change referred to as *decoherence*. In some situations it is possible to find a subspace of the $N$-dimensional Hilbert space which is unaffected by specific types of decoherence. This is a *decoherence-free subspace* (DFS) [Zan97, Lid98, Dua98]. For the three-state system probability loss from state 2 (only) is a form of decoherence that does not affect the dark state: it is a simple two-dimensional example of a DFS for decoherence that originates with population loss.

### 1.3.12 Photons

In setting up the RWA Hamiltonian for STIRAP in **Sec.** 1.2.1 I used what is usually called the semiclassical approach to quantum-state changes, an approach in which the fields have so many photons that a change by one is not noticeable and the fields can therefore are treated as given functions of time, unaffected by the excitation. It is that view that underlies, for example, the discussion of Autler-Townes splitting in **Sec.** 2.4.

The two fields of STIRAP may be those of traveling waves, such as a laser beam, or they may be those of a standing-wave cavity field in which the discreteness of photon numbers, $n_P$ and $n_S$, must be treated. When treating quantized fields and definite numbers of photons the basis states then include descriptors of atom states, $P$-field states and $S$-field states, as in the use of photon-number states in the replacements (see [Wal70, Wal71b, Rad82, Yoo85]):

$$|1\rangle \to |1, n_P, n_S\rangle, \qquad |2\rangle \to |2, n_P - 1, n_S\rangle, \qquad |3\rangle \to |3, n_P - 1, n_S + 1\rangle. \tag{1.41}$$

With this quantized-field description the STIRAP transition from the initial atom state 1 to the final state 3 removes one $P$ photon and adds one $S$ photon. The reverse process replaces an $S$ photon by a $P$ photon. The RWA amounts to considering only the three energy-conserving states above, although there exist other links that do not conserve energy.

The equations also obtain by Floquet treatment of harmonically varying constant-intensity classical fields [Shi65, Gue97]. For excitation by laser beams the photon numbers $n_P$ and $n_S$ are so large that their quantum properties are not observable, and a classical description is satisfactory.

The Rabi frequency for a field of $n$ photons is $\Omega_{vac}\sqrt{n+1}$ where $\Omega_{vac}$ is the *vacuum Rabi frequency*. For the usual optical transitions the Rabi frequency expresses an electric-dipole interaction energy and so $\hbar|\Omega_{vac}| = |d_{12}\mathcal{E}_{vac}|$ where $d_{12}$ is the electric-dipole transition moment and $\mathcal{E}_{vac}$ is the single-photon electric-field amplitude. With a cavity of frequency $\omega_c$ and volume $V_c$ this has the magnitude [Har89] $|\mathcal{E}_{vac}| = \sqrt{\hbar\omega_c/(2\epsilon_0 V_c)}$.

### 1.3.13 Pulse-shape insensitivity

Adiabatic processes typically have the property that the final state of the system does not depend on details of the time variation of the Hamiltonian. For pulsed coherent excitation this means that details of pulse shapes, and of frequency variation, are not important. In this respect adiabatic

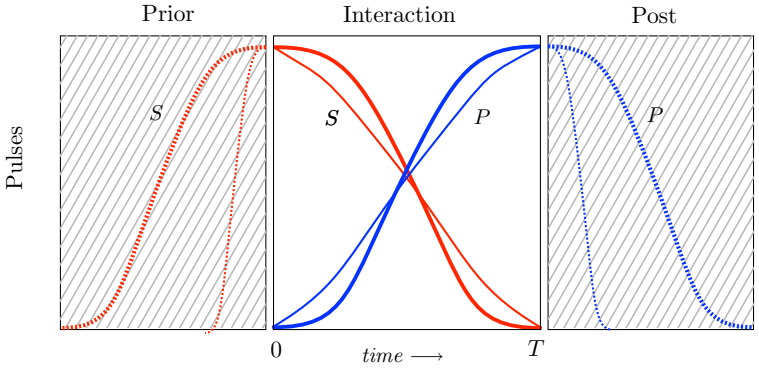


Fig. 1.9. Examples of pulse envelopes of finite duration $T$, all of which can accomplish STIRAP. Dotted lines show envelope portions that are immaterial. Thick and thin lines show alternative pulse shapes. The thinner lines, full and dashed, are what might be termed "shark-fin" pulses. The thicker lines are distortions of hypergaussian shapes, $\exp[-(t/\tau)^n]$, with $n > 2$.

dynamics differs significantly from resonant population transfer induced by pi pulses [Rab54, All75,Sho90,Vit01a,Vit01b,Sho11], that produce desired results by carefully controlled temporal pulse areas.

**Figure** 1.9 illustrates an aspect of this insensitivity: Prior to the arrival of the $P$ pulse the $S$ envelope-shape is immaterial; cf eqn. (G.22). Following the termination of the $S$ pulse the $P$ envelope is immaterial. The prior and post intervals, shown shaded in the figure, have no effect on the mixing angle that determines the structure of the dark state.

Although the success of STIRAP does not depend on any details of the pulse envelope, a choice of pulse shape, for fixed laser power or temporal pulse area, can effect the adiabaticity [Gue02, Tor08]. In practice, this means that for a given pulse area or peak Rabi frequency the nonadiabaticity error can be minimized by suitable choice of pulse envelope.

### 1.3.14 Degeneracy treatment

Unlike the rate equations that describe the absorption, stimulated emission and spontaneous emission of Einstein's description of radiative processes (cf. **App.** D), the TDSE does not deal with sets of states as a single unit; it deals with individual quantum-state probability amplitudes. To treat degenerate states, for example an initial situation in which there is angular momentum degeneracy (i.e. quantum states are *Zeeman sublevels*), it is necessary to include each quantum state individually and explicitly in the description, thereby enlarging the Hamiltonian matrix: a level having angular-momentum quantum number $J$ has $2J + 1$ distinguishable sublevels and hence it requires $2J + 1$ quantum states for its description; see [Zar88].

With allowance for angular momentum the linkage pattern of the Hamiltonian matrix depends on the polarizations of the $P$ and $S$ fields [Zar88, Sho90, Sho11]. When the field polarizations (the unit vectors $\mathbf{e}_P$ and $\mathbf{e}_S$) are chosen appropriately, the linkage pattern of a multistate system may be reducible to a three-state chain. **Figure** 1.10 shows two examples, in which $\mathbf{e}_P$ and

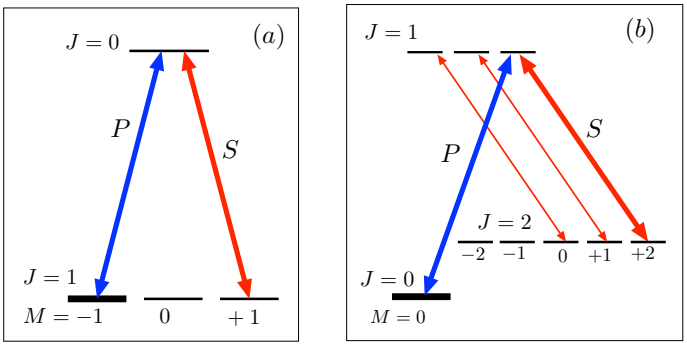


Fig. 1.10. $(a)$ A stimulated Raman linkage between degenerate Zeeman sublevels of a two-level atom, using $P$ and $S$ fields of opposite circular polarization. STIRAP moves between degenerate sub levels $M = -1$ and $M = +1$ of $J = 1$. $(b)$ A stimulated Raman linkage between degenerate Zeeman sublevels of a three-level atom. STIRAP will selectively excited a single sublevel, $M = +2$ of $J = 2$, starting from $M = 0$ of $J = 0$. Lighter $S$ lines are unconnected to the three-state $P - S$ linkage and do not affect the STIRAP process.

$\mathbf{e}_S$ describe opposite circular polarization. In the example of frame $(a)$ a crafted time-variation of single-field polarization, from $\mathbf{e}_S$ to $\mathbf{e}_P$, will implement STIRAP between two degenerate sublevels. In frame $(b)$ amplitude-varying STIRAP will transfer population into a single sublevel.

### 1.3.15 Averages

The TDSE applies to individual quantum systems, e.g. single isolated atoms or molecules. Experiments generally deal with ensembles of individual systems, each distinguished by some attribute $e$ such as center-of-mass motion, initial orientation, initial excitation energy or neighboring particles. Each individual system obeys an equation of the form

$$\mathrm{i}\frac{d}{dt}\mathbf{C}(e;t) = \mathsf{W}(e;t)\mathbf{C}(e;t). \tag{1.42}$$

What is experimentally observable is the average over the ensemble, for example the population at time $t$. This is evaluated by summing the populations for each environment,

$$P_n(t) = \sum_e p(e)\,|C_n(e;t)|^2, \tag{1.43}$$

weighted by $p(e)$, the probability of finding environment $e$.

It is generally not possible to find an "average atom" Hamiltonian $\bar{\mathsf{W}}(t)$ such that its probability amplitudes $\bar{C}_n(t)$,

$$\mathrm{i}\frac{d}{dt}\bar{\mathbf{C}}(t) = \bar{\mathsf{W}}(t)\bar{\mathbf{C}}(t), \tag{1.44}$$

give the required average probabilities

$$P_n(t) = |\bar{C}_n(t)|^2. \tag{1.45}$$

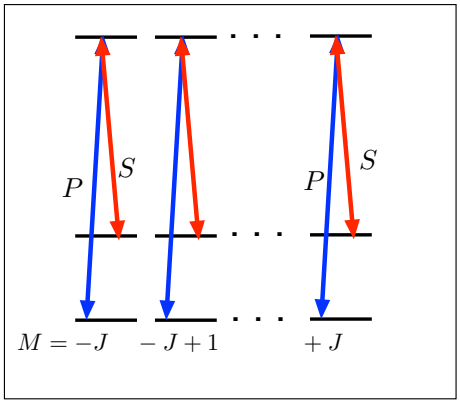


Fig. 1.11. Contributing subsystems for STIRAP with degenerate Zeeman sublevels. Each chain has a distinct set of $M$-dependent dipole moments and hence observations of population dynamics will average over different Rabi frequencies.

However, adiabatic following offers an exception. The first STIRAP experiments [Gau88,Gau90] dealt with rotational states of molecules, i.e. a system with degenerate Zeeman sublevels. The two laser fields were linearly polarized and so the system could be described by a set of independent three-state STIRAP equations, each labeled by the magnetic quantum number of the ground state, as in **Fig.** 1.11; see also **App.** E.3. By ensuring that the subsystem with weakest links underwent STIRAP the experimenters induced population transfer of all the linked sublevels ($M = 0$ is unlinked). Their published results are thus of measured sublevel averages.

## 2 The pre-history of STIRAP

The preceding subsections list characteristics that are associated with what is here defined as STIRAP. For a process to be labeled an example of STIRAP these should be observable. Each of them has had an independent history of study.

The following major portion of this review discusses some of the many articles that present aspects of the theory underlying STIRAP. These deal with a variety of topics, primarily in atomic, molecular and optical physics, that are part of the broadly developing physics of STIRAP, most of which were mentioned in the preceding section as characteristic ingredients of STIRAP.

### 2.1 Basic background

The notion of stationary states of electron motion within atoms, and of transitions gaining and losing discrete electromagnetic energy increments, date from the papers of Bohr [Boh13] and Einstein [Ein16] that set the stage for modern quantum theory.

#### 2.1.1 Quantum states

Because STIRAP was developed as a procedure for producing changes to a quantum-mechanical system, its origins trace to the works in the 1920s that created the present quantum theory, with its concepts of discrete quantum states and the time-dependent Schrödinger equation (TDSE), as is discussed in [van68] and numerous textbooks, e.g. [Sch55, Dir58, Mes62].

The steps leading, by means of the statevector expansion (1.5), from the TDSE (1.9) to coupled ODEs as in eqn. (1.10), is often attributed to Dirac [Dir26] as the "variation of constants" method. It has counterparts in variational methods, such as Rayleigh-Ritz-Galerkin methods [Sho73a, Sho73b], and finite-element methods, for solving partial differential equations [Bur87, Joh87], all of which rely on basis vectors in a multidimensional abstract vector space.

#### 2.1.2 Adiabatic change, 1916, 1928

Adiabatic change is key to the functioning of STIRAP. In thermodynamics an adiabatic process is one that involves no transfer of heat or matter between a system and its surroundings. Notions of adiabatic change in quantum theory trace to work of Ehrenfest [Ehr16a, Ehr16b] predating modern quantum theory and the Schrödinger equation. A 1928 paper by Born and Fock [Bor28a] introduced the idea of a system adapting its quantum state to gradually changing conditions (the *adiabatic theorem*).

#### 2.1.3 The LZSM papers, 1932

STIRAP relies on adiabatic change of the coefficients in the TDSE, for which there are early predecessors applied to two quantum states and associated with the names of Landau [Lan32a, Lan32b], Zener [Zen32], Stückelberg [Stu32] and Majorana [Maj32]; see [Gia05]. Historically, such change was induced by a monotonically changing (sweeping) energy (a chirped detuning) and is often given the acronym LZ or LZS or LZSM. Those works, and subsequent interest,

cf. [Nik99], dealt with slowly changing quasistatic fields, such as occur between colliding atoms or molecules.

The LZSM papers were also significant for providing analytic solutions to the TDSE, a notable feature found also in STIRAP. In the LZSM model the Rabi frequency was constant and the detuning a linear function of time, giving parabolic cylinder functions as the solutions to the two-state TDSE.

### 2.2 Vector pictures and torque equations, 1946–84

The excitation probabilities $P_n(t)$ provide only a portion of the information present in a statevector and its Hilbert-space components. A number of ways of organizing and depicting additional information have been useful in descriptions of coherent excitation. This section discuss some of them that deal with abstract vectors. Their equations of motion, treated as torque equations, provide a very simple picture of adiabatic change as adiabatic following — the continuing alignment of two abstract vectors. As noted in **Sec.** 1.3.5, the interpretation of the TDSE for STIRAP as a torque equation makes clear the characteristics of STIRAP dynamics.

#### 2.2.1 The Bloch equations, 1946

The development of magnetic resonance techniques led to the use of radio-frequency (rf) fields to induce adiabatic changes between two states [Blo46, Tow55, Abr61]. Writing in 1946 a review of magnetic resonance [Blo46] Bloch derived a torque equation for the collective magnetic moment $\mathbf{M}$ of an ensemble of nuclear spins in the presence of a magnetic field $\mathbf{H}$:

$$\frac{d}{dt}\mathbf{M} = \gamma \mathbf{M} \times \mathbf{H}, \tag{2.1}$$

where $\gamma$ is the gyromagnetic ratio (of magnetic moment to angular momentum). Bloch took particular interest in a magnetic field that had a static longitudinal component $H_0$ and a transverse component that varied at the rf frequency $\omega$ close to the resonance frequency $\omega_0 = \gamma H_0$:

$$H_x = H_1 \cos \omega t, \qquad H_y = \mp H_1 \sin \omega t, \qquad H_z = H_0, \tag{2.2}$$

(the signs refer to positive and negative values of $\gamma$) and, defining a resonance field $H^* = \omega/|\gamma|$ and detuning $\delta = (H_0 - H^*)/H_1$ he obtained a solution for $\mathbf{M}(t)$ that rotated around the $z$ axis.

Bloch noted the condition for adiabatic change, either by varying the magnitude of the $z$-component $H_0$ or varying the frequency of the rf field $\omega$, was $|d\delta/dt| \ll |\gamma H_1|$.

**Relaxation times $T_1$ and $T_2$.** Bloch treated incoherent effects by introducing time constants $T_1$ and $T_2$ that characterized expected rate-equation relaxation of the oscillating magnetic moment $\mathbf{M}$: the relaxation of the "longitudinal" component $M_z$ was determined by $T_1$, while $T_2$ governed the "transverse" components $M_x$ and $M_y$. Using these he gave the differential equation for the three component of $\mathbf{M}$ as what are now known as the *Bloch equations*, written here in vector-matrix form:

$$\frac{d}{dt}\begin{bmatrix} M_x \\ M_y \\ M_z \end{bmatrix} = \begin{bmatrix} (1/T_2) & -\gamma H_z & \gamma H_y \\ \gamma H_z & (1/T_2) & -\gamma H_x \\ -\gamma H_y & \gamma H_x & (1/T_2) \end{bmatrix} \begin{bmatrix} M_x \\ M_y \\ M_z \end{bmatrix} + \frac{1}{T_1}\begin{bmatrix} 0 \\ 0 \\ M_{eq} \end{bmatrix}, \tag{2.3}$$

where $M_{eq} = \chi H_0$ is the equilibrium magnetization ($\chi$ is the nuclear paramagnetic susceptibility).

**Bloch variables $u$ and $v$.** Bloch made use of coordinates that rotated with the field frequency, defining slowly-varying (Bloch) variables $u$ and $v$ through

$$M_x = u\cos\omega t - v\sin\omega t, \qquad M_y = \mp(u\sin\omega t + v\cos\omega t). \tag{2.4}$$

The Bloch equations became the basis for rapidly growing work on nuclear magnetic resonance (NMR) [Abr61, Sli63], and then, with replacement of magnetization $\mathbf{M}$ by an electric dipole moment, a foundation for describing laser-induced changes [All75,Mil76,Fen77,Sho90,Coh08]. The earliest applicable equations were those described in **Sec.** 2.2.2 below.

### 2.2.2 The Feynman-Vernon-Hellwarth torque equation, 1957–64

In 1957 Feynman, Vernon and Hellwarth (FVH) [Fey57] presented a transcription of the density-matrix equations for coherent two-state excitation into the form of a three-dimensional torque equation. Written with notation appropriate for the present article the equation reads (with suppression of time arguments)

$$\frac{d}{dt}\mathbf{r} = \boldsymbol{\Upsilon}\times\mathbf{r}, \tag{2.5}$$

where the three components of the unit vector $\mathbf{r}$ are formed from bilinear products of probability amplitudes, in the present notation

$$r_1 \equiv u = C_2C_1^* + C_1C_2^*, \qquad r_2 \equiv v = \mathrm{i}[C_2C_1^* - C_1C_2^*], \qquad r_3 \equiv w = C_2C_2^* - C_1C_1^*. \tag{2.6}$$

The vector $\mathbf{r}$, of constant unit length, is known as the *Bloch vector* and its motion takes place on the *Bloch sphere*. Components $r_1$ and $r_2$ are termed *coherences*; component $r_3$ is the *population inversion.* The elements of the angular-velocity vector $\boldsymbol{\Upsilon}$ (also known as "the torque vector") are taken from the RWA Hamiltonian W, with allowance for possible complex values,

$$\Upsilon_1 = W_{21} + W_{12}, \qquad \Upsilon_2 = \mathrm{i}[W_{21} - W_{12}], \qquad \Upsilon_3 = \Delta. \tag{2.7}$$

A simple example of a two-state quantum system is an angular momentum (or spin) $J = \frac{1}{2}$, and the terminology of such a system (as a "pseudo spin") is often used in discussions of two-state dynamics, whatever the physical system may be. Although the dominant interest of researchers remained two-state transitions, as fitted a system of spin one-half, in 1964 Morris extended that work to treat the three states of a spin-one systems [Mor64]. He presented a torque equation akin to that of FVH, anticipating work done later, cf. **Sec.** 2.2.8. As with earlier work, this considered swept frequencies as the adiabatically-varying contributions to the Hamiltonian.

### 2.2.3 Adiabatic following, 1964

With the availability of lasers interest grew in schemes to use these as the interaction that would induce two-state adiabatic transitions. Abella, Kurnit and Hartmann [Abe64] applied the FVH

vector model to describing photon echoes and presented equations for two-state coherent excitation expressed as a torque equation for a "pseudo-electric dipole moment", an electric-dipole counterpart of the magnetic moment $\mathbf{M}$ treated by Bloch [Blo46], cf. **Sec.** 2.2.1. This was used as the basis for work by experimental work Grischkowsky [Gri70] who said: "the pseudomoment will remain aligned along the changing effective field" (meaning the angular-velocity vector $\boldsymbol{\Upsilon}$). Grischkowsky and Armstrong [Gri71] refer to the "adiabatic following model" of [Gri70] as a contrast to the steady-state models of others. They say "the term 'adiabatic following' describes the situation in which the pseudomoment of the near-resonant transition follows (remains parallel to) the effective field of the laser pulse." Other publications using this model soon appeared [Cri73, Gri73, Gri75].

Basically, as noted in **Sec.** 1.3.5, given a torque equation in the form (2.5) or (1.30), the two vectors $\mathbf{r}$ and $\boldsymbol{\Upsilon}$ will remain aligned if the variation of $\boldsymbol{\Upsilon}$ is slow and the two vectors are either parallel or antiparallel. so that $\boldsymbol{\Upsilon} \times \mathbf{r} = 0$. Those are the conditions for adiabatic following (of $\boldsymbol{\Upsilon}$ by $\mathbf{r}$).

### 2.2.4 Two-photon vector models, 1971–75

The FVH vector model was extended to two-photon transitions by Takatsuji [Tak71], who dealt with large detuning and the effective two-state Hamiltonian obtained by adiabatic elimination (see **Sec.** 2.10.1). In this revision the Rabi frequency is proportional to the product of two field envelopes and the detuning includes dynamic Stark shifts. This work was extended by Grischkowsky, Loy and Liao [Gri75] to allow two independent fields. They described situations in which a Bloch vector remained aligned with (adiabatically followed) an angular-velocity vector.

In 1975 Brewer and Hahn [Bre75], treating single-field excitation of a three-state system subject to two-photon resonance, introduced the variables

$$u_{ij} = \rho_{ij} + \rho_{ji}, \qquad \mathrm{i}v_{ij} = \rho_{ij} - \rho_{ji}, \qquad w_{ij} = \rho_{ii} - \rho_{jj}, \tag{2.8}$$

the single-photon detuning

$$\Delta = \omega - \omega_{21} = \omega - \omega_{32}, \tag{2.9}$$

and the half Rabi frequencies

$$\alpha = \tfrac{1}{2} d_{13}\mathcal{E}/\hbar, \qquad \beta = \tfrac{1}{2} d_{23}\mathcal{E}/\hbar. \tag{2.10}$$

From these they defined the three variables

$$U = (\alpha u_{13} + \beta u_{23})/\Omega, \tag{2.11a}$$

$$V = (\alpha v_{13} + \beta v_{23})/\Omega, \tag{2.11b}$$

$$W = 2(\alpha^2 w_{13} + \beta^2 w_{23} + \alpha\beta u_{12})/\Omega^2, \tag{2.11c}$$

where $\Omega = 2\sqrt{\alpha^2 + \beta^2}$ is the rms Rabi frequency of the two fields. As they stated, these variables satisfy an equation analogous to the torque equation of FVH:

$$\frac{d}{dt}\begin{bmatrix} U \\ V \\ W \end{bmatrix} = \begin{bmatrix} 0 & \Delta & 0 \\ -\Delta & 0 & \Omega \\ 0 & -\Omega & 0 \end{bmatrix} \begin{bmatrix} U \\ V \\ W \end{bmatrix}. \tag{2.12}$$

Brewer and Hahn were concerned with propagation effects, and considered both the lambda and ladder linkages. They were interested in steady-state solutions and decaying pulsed excitation, when $T_1$ and $T_2$ relaxation times had to be considered. They did not consider adiabatic changes nor did they point out population trapping.

### 2.2.5 Adiabatic rapid passage, 1974–76

Loy, in 1974 [Loy74] observed population inversion attributed to optical *adiabatic rapid passage* (ARP). The main requirements, as he noted, were that either the frequency of the two-level system or that of the laser must be swept through the linewidth of the system in a time short compared to the relaxation time and that the laser field must be sufficiently strong that the time evolution is adiabatic.

In 1975 Grischkowsky, Loy and Liao [Gri75] treated adiabatic following for two-photon transitions. This was discussed by Loy and Grischkowsky [Loy76, Loy77]. In these various works the adiabatic changes were produced by swept frequencies, as were those discussed in the text by Allen and Eberly [All75]. These processes, which relied on a linear sweep of detuning in the two-state version of eqn. (1.10), were termed variously *adiabatic following* (AF) and *rapid adiabatic passage* (RAP) or *adiabatic rapid passage* (ARP), with the adjective "rapid" indicating that the transitions occurred on a time scale much shorter than spontaneous-emission lifetimes.

The noun "following" originally made reference to the alignment of the Bloch vector with the controlled angular-velocity (torque) vector. In the context of STIRAP it usually refers to alignment of the statevector with an adiabatic state; but see **Sec.** 1.3.5. The noun "passage" historically restricted consideration to situations in which there occurs completed population transfer.

### 2.2.6 Adiabatic passage with decoherence, 1982–84

A somewhat different approach to adiabatic passage, suiting a view of laser-induced excitation common at the time, was considered by Peterson and Cantrell [Pet82, Pet85]. They were interested in exciting molecules whose electronic excitation could be modeled as a succession of bands of closely-spaced energy levels, to be excited by a set of simultaneous near-monochromatic pulses, as shown in **Fig.** 2.1. They considered adiabatic evolution of dressed states to accomplish population transfer, noting that (for their system) adiabatic changes would be reversible (but see **Sec.** 1.3.7) so that to produce permanent population transfer they required decoherence-inducing processes, i.e. the procedure was, in part, adiabatic but not rapid.

### 2.2.7 Generalized Bloch equation, 1980

A paper by Elgin in 1980 [Elg80] generalized the three-dimensional Bloch-vector description of two-state dynamics introduced by FVH [Fey57] to the three-state chain excited by two electromagnetic pulses by means of an eight-component vector $\mathbf{r}$. He used the generators of the SU(3) group to express both the RWA Hamiltonian and the density matrix, thereby deriving a torque equation for the dynamics in the absence of relaxation, and making evident the rotational nature of the statevector: the generators of this statevector rotation are the generators of the SU(3) transformation group. Elgin pointed out constants of the motion but gave no illustrative examples.

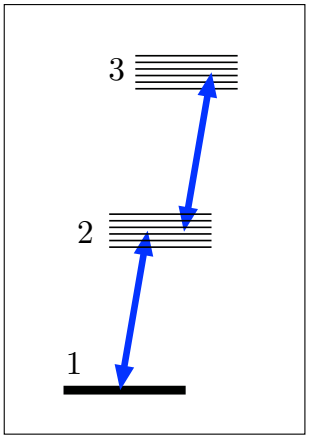


Fig. 2.1. Schematic of excitation into bands of levels, as considered by [Pet82, Pet85]. Redrawn based on Fig. 1 of [Pet82]. Reprinted from [Pet82] with permission from Elsevier.

### 2.2.8 The coherence vector, 1981–84

A series of papers by Hioe, some with colleagues Eberly and Carroll [Hio81, Hio82a, Hio82b, Hio82c, Hio83a, Hio83b, Hio84a, Hio84b], and Oreg [Ore84] introduced a formalism for treating coherent $N$-state quantum dynamics by means of a *coherence vector* $\mathbf{S}(t)$ of dimension $N^2-1$ and unit length, whose real-valued elements, generalizing the three components of the Bloch pseudo spin, are obtained from elements of the $N$-state density matrix. From the bilinear products

$$u_{jk}=\rho_{jk}+\rho_{kj},\quad v_{jk}=-\mathrm{i}[\rho_{jk}-\rho_{kj}],\quad w_l=-\sqrt{2/l(l+1)}[\rho_{11},\cdots,\rho_{ll}-l\rho_{l+1,l+1}], \tag{2.13}$$

they formed an ordered array, a vector of constant length

$$\mathbf{S}=\{u_{12},\ldots,v_{12},\ldots,w_1,\ldots,w_{N-1}\}, \tag{2.14}$$

that satisfies a multidimensional generalization of the three-dimensional torque equation presented by FHV [Fey57] and thereby undergoes rotation in $N^2-1$ dimensions. In their words the coherence vector extends the Bloch or pseudo-spin vector to $N$ states interacting with external forces of "arbitrary strength, time dependence and resonance character". They pointed out that products of $n$ density matrices of an $N$-state system,

$$c(N,n)=\mathrm{Tr}\,[\rho(t)^n],\quad n=1,2,\ldots,N, \tag{2.15}$$

were constants of the motion, an extension of earlier results by Elgin [Elg80] for $N=3$.

The components of the coherence vector can, more generally, be defined by

$$S_j(t)=\mathrm{Tr}\,[\rho(t)\mathsf{s}_j], \tag{2.16}$$

where the $\mathsf{s}_j$ are generators of the SU(N) group. Choices of group representations, fitting the symmetry of the Hamiltonian linkage, allow simplification of the description of $\mathbf{S}(t)$ [Hio83b, Hio85a, Hio87, Hio88a, Hio88b, Hio89]. Hioe and Eberly [Hio82a] showed that, under appropriate conditions, the eight-dimensional space of the coherence vector for a three-state system can be decomposed into three independent subspaces, of dimension one, three and four, with three independent coherence vectors. The squares of the lengths of these vectors are constants of the motion. These results were presented, summarily, in [Hio82b]. Hioe has written a detailed review of these and related results of dynamic symmetries [Hio09].

In 1985 Ho and Chu [Ho85] gave examples, with illustrations, of the oscillatory motion of the $N = 3$ coherence vector for steady monochromatic fields.

### 2.2.9 The coherence vector and adiabatic following, 1983–84

The 1982 work of Hioe and Eberly [Hio82a] applied this formalism to the lossless three-state chain linkage at exact two-photon resonance, allowing single-photon detuning, with identical time dependence $f(t)$ for $P$ and $S$ pulses. The 1983 Hioe paper [Hio83a] showed numerical results for linearly varying single-photon detuning, a three-state version of the established two-state adiabatic passage. Hioe and Eberly [Hio84a] examined this model, of simultaneous pulses, for the effects of various analytic expressions for pulse shapes and detunings. They used sech pulses and tanh detuning variation, as were used by Allen and Eberly for two states [All75], to obtain analytic solutions for elements of the density matrix. Further papers discussed the extension of the two-state LZSM theory of adiabatic passage (see **Sec.** 2.1.3) to three states and two simultaneous pulses [Car86a, Car86b].

Although this example was one of three-state adiabatic passage, the conditions were not those of STIRAP: adiabatic passage was driven by chirped frequencies rather than pulse amplitude variations, and population passed through the intermediate state 2.

The 1984 paper of Oreg, Hioe and Eberly [Ore84] extended preceding discussion of the $N$-state coherence vector $\mathbf{S}(t)$ stressing its use as a general formalism for treating multi-state dynamics, and adiabatic following in particular. With suitably defined generalization of the torque vector of the Bloch equation, adiabatic motion can be viewed as the vector $\mathbf{S}(t)$ following (i.e. remaining aligned with) the torque vector.

As stated in its abstract, this paper paid specific attention to the production of complete population inversion in a chain linkage whose interaction was by "a continuous sweep of the laser field frequencies and/or envelopes". The word *sweep* used here, and elsewhere in the article, suggests that they have in mind a chirped-frequency procedure, as in earlier work on two-state adiabatic passage. Indeed that is what they discuss in their specific example (from [Hio83a]) — two chirped laser frequencies — but the coherence-vector formalism also fits alternative procedures in which the frequencies are constant. Their stated interest was in finding a set of detunings and Rabi frequencies such that by "sweeping" them adiabatically from initial to final values the coherence vector will be led from some initial $\mathbf{S}(0)$ to some prescribed final $\mathbf{S}(T)$.

A narrow interpretation of this paper, based on their examples, is that they have in mind chirping of frequencies. However, a sweep of Rabi frequencies is also a possibility they considered (see **Fig.** 2.2), and if these are swept "counter-intuitively", as was done later with "shark-fin" pulses [Car92, Yat98a, Tor08], then the result is the traditional STIRAP pulse sequence. That notion is not explicitly recognized in the paper [Ore84] and there is no indication that the authors

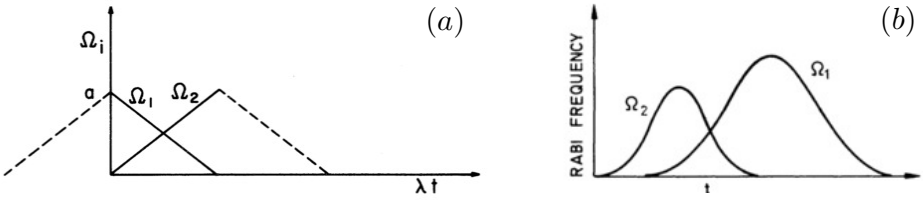


Fig. 2.2. $(a)$ Suggestion by Oreg, Hioe and Eberly [Ore84] for pulses that will accomplish three-state adiabatic passage when single-photon detuning $\Delta$ is very large and the system behaves as two states. Here $\Omega_1$ (the $P$ field) precedes $\Omega_2$ (the $S$ field), the intuitive ordering, and $\lambda$ is the linear chirp rate used in modeling swept detuning. From Fig. 7 of [Ore84]. Reprinted with permission from [Ore84]. Copyright 1984 by the American Physical Society. $(b)$ The counter-intuitive pulse sequence used by Kuklinski, Gaubatz, Hioe and Bergmann [Kuk89] and shown as their Fig. 2. The $S$ field, here $\Omega_2$, precedes the $P$ field. Reprinted with permission from [Kuk89]. Copyright 1989 by the American Physical Society.

examined this possibility other than with very large single-photon detuning and the resulting two-state simplification; see **Sec.** 2.10.1.

After their eqn. (39) the authors say conditions are "anti-intuitive": they speak of not following the "natural" sequence of 1-2 excitation and then 2-3 excitation, but instead using "something close to the opposite order of resonances" when moving population from state 1 to state 3. But these are "anti-intuitive" conditions on detunings and chirps, not on the timing of pulsed Rabi frequencies. Their example, starting with their eqn. (42), treats simultaneous $P$ and $S$ pulses that have different detunings: the 2-3 resonance occurs first, followed by the 1-2 resonance, and they do not maintain two-photon resonance. So this is not the counter-intuitive aspect traditionally associated with STIRAP [Sho95b].

Their illustration of population histories, shown in their Fig. 4 taken from Fig. 1 of [Hio83a] (see **Fig.** 6.3), shows appreciable population in the intermediate state. So this is not an example of population transfer using the dark state of STIRAP. Their Fig. 3, from [Hio83a], shows the adiabatic eigenvalues. There is no constant value, so again this is seen not to be STIRAP, although it is certainly adiabatic following.

Their Fig. 7 [shown here as frame $(a)$ of **Fig.** 2.2] shows sequential pulses, but it is the $P$ pulse, coupling states 1 and 2, that occurs first. This is not the $S$-first STIRAP sequence[13]. However, it seems clear that their figure was intended merely to support their suggestion that properly timed pulses could produce complete population transfer: their accompanying formulas for pulse shapes describe descending $S$ field and rising $P$ field, as occurs with counter-intuitive pulse ordering. They gave no illustrations of population histories with delayed pulses; their computations were for the limiting case of large single-photon detuning, used with adiabatic elimination and consequent two-state dynamics; see **Sec.** 2.10.1.

**Summary: Coherence vector.** The development of the coherence vector showed how one could generalize the two-state Bloch vector to a multidimensional vector that satisfied a torque-

[13]Their figure shows $P$-before-$S$ but the formulas of their figure caption, and in their text, are for $S$-before-$P$.

like equation, and could from this find a vector that was unaffected by the moving torque — how one could find Hilbert space vectors that underwent adiabatic following. But although this paper and [Hio83a] provided a useful formalism for treating adiabatic following, including STIRAP, the paper does not mention key conditions that differentiate STIRAP from other forms of AF. Their illustrative examples produced complete population transfer, i.e. were AP, but were not STIRAP. Their focus was on the coherence vector, not on adiabatic states, as a means of picturing adiabatic following.

## 2.3 Population trapping, 1951–1967

The dynamic use of a dark, population-trapping state is an essential ingredient of STIRAP, distinguishing it from other forms of adiabatic following. The 1951 Lamb paper [Lam51] described a means of preserving population in a three-state system in the presence of quenching effects. This section discusses notions described by Lamb and his co-workers that are identifiable as elements of STIRAP. Missing from the Lamb work is the adiabatic following needed for STIRAP.

### 2.3.1 The Lamb papers: Population trapping, 1950–52

In a series of papers starting in 1950 Lamb and Retherford discussed population changes in hydrogen. The presented theory dealt with a lambda linkage and adiabatic eigenstates. The first three papers, discussed below, have relevance to STIRAP. Further publications presented data on the fine structure of hydrogen and deuterium, but are not relevant to STIRAP.

### 2.3.2 First Lamb paper, 1950

A 1950 paper of Lamb and Retherford [Lam50] began the series of publications on the fine structure of the hydrogen atom. It provided needed background and notational definitions but it did not treat excitation dynamics.

The paper presented a diagram of the structure of the 2S and 2P energy levels [14] of hydrogen in a magnetic field, with neglect of nuclear spin,. The 2P states $(a, b, c, d, e, f)$ undergo spontaneous emission to the 1S states, whereas the 2S states $(\alpha, \beta)$ are metastable: their loss takes place by two-photon decay, by collisions or by transition into 2P states induced by electric fields. The authors noted that when a magnetic field $\mathbf{B}$ is present, atom motion with velocity $\mathbf{v}$ introduces an electric field proportional to $\mathbf{v} \times \mathbf{B}$ that can cause such transitions.

### 2.3.3 Second Lamb paper, 1951

In the second paper by these authors [Lam51] there was discussion of two specific radio-frequency transitions, shown as $M$ and $R$ in **Fig.** 2.3(b). Again neglecting nuclear spin, one can say that the $M$ transition is between two Zeeman sublevels of the 2S configuration, transition $\alpha - \beta$, while the other transition is between sublevels of the 2S and 2P configuration, specifically the transition $\alpha - e$. The regime of interest is when the sublevels $\beta$ and $e$ are resonant, or nearly resonant, a region shown in frame $(a)$ with a yellow cartouche. In this region only three Zeeman sublevels

[14] Traditionally the quantum labels of a single electron would be denoted $2s$ and $2p$, with lower case letters denoting orbital angular momentum. Here I follow the notation of Lamb and use upper case letters.

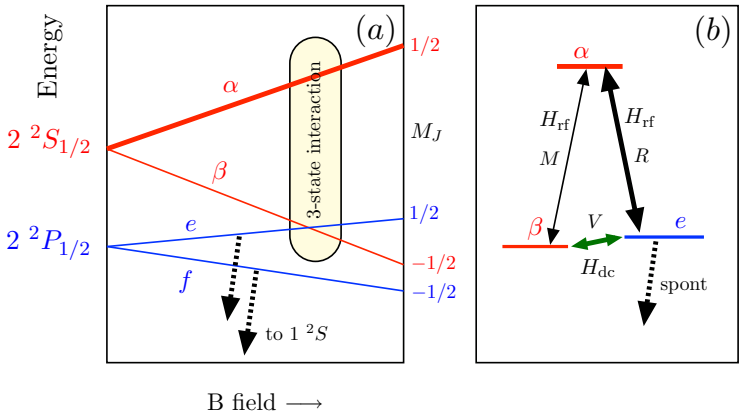


Fig. 2.3. Left: $(a)$ Zeeman sublevels of hydrogen involved in three-state dynamics, with neglect of nuclear spin. A cartouche marks the region of interest, shown in frame $(b)$. Redrawn after Fig. 11 of [Lam50]. Reprinted with permission from [Lam50]. Copyright 1950 by the American Physical Society. $(b)$ Linkage pattern for interactions considered in [Lam51, Lam52] and [Ohl67]. There are two interactions at radio-frequency (rf) $\omega$, with strengths $M$ (magnetic dipole) and $R$ (electric dipole), and a quasi-static electric interaction with strength $V$, in the notation of [Lam51, Lam52, Ohl67].

(three quantum states) need be considered, namely the metastable states $\alpha, \beta$ and the decaying state $e$:

$$\alpha: (2\,^2S_{1/2}, m_s = \tfrac{1}{2}), \quad \beta: (2\,^2S_{1/2}, m_s = -\tfrac{1}{2}), \quad e: (2\,^2P_{1/2}, m_j = \tfrac{1}{2}). \tag{2.17}$$

The Hamiltonian linkages of these quantum states are shown in the frame at the right. There are two steady radio-frequency (rf) fields, and a static (dc) electric field that, together, form a triangle linkage pattern. In the notation of [Lam51, Lam52] and [Ohl67] these have interaction strengths $R$, $M$, and $V$ respectively. The $M$ interaction is of magnetic dipole, the $R$ interaction is of electric dipole, and the $V$ interaction is quasi-static.

### 2.3.4 Third Lamb paper, 1952

The third paper of the set, by Lamb alone [Lam52], considered in great detail the energy structure of hydrogen, taking into account various relativistic effects of the electron-nucleus interaction, including hyperfine structure, and the effects of externally applied electric and magnetic fields. It paid particular attention to the quantum states — Zeeman sublevels — that appear in **Fig.** 2.3. Lamb considered a three-state system in the RWA and interaction picture (see **Sec.** C) for the states $\alpha, \beta, e$. He presented, in his eqn. (207), a time-dependent Schrödinger equation that can be written (with his notation for elements of the Hamiltonian but reordered statevector components)

$$\mathrm{i}\frac{d}{dt}\begin{bmatrix} C_\alpha \\ C_e \\ C_\beta \end{bmatrix} = \frac{1}{2}\begin{bmatrix} 0 & R^*\mathrm{e}^{\mathrm{i}(\delta+\omega_{\beta e})t} & M^*\mathrm{e}^{\mathrm{i}\delta t} \\ R\mathrm{e}^{-\mathrm{i}(\delta+\omega_{\beta e})t} & -\mathrm{i}\gamma & 2V\mathrm{e}^{-\mathrm{i}\omega_{\beta e}t} \\ M\mathrm{e}^{-\mathrm{i}\delta t} & 2V^*\mathrm{e}^{\mathrm{i}\omega_{\beta e}t} & 0 \end{bmatrix}\begin{bmatrix} C_\alpha \\ C_e \\ C_\beta \end{bmatrix}, \tag{2.18}$$

where the rf frequency of the two interactions is $\omega$ and the two Bohr frequencies are $\omega_{\alpha\beta}$ and $\omega_{\alpha e}$,

$$\hbar\omega_{ij} = |E_i - E_j|, \qquad \delta = \omega_{\alpha\beta} - \omega. \tag{2.19}$$

**Eigenstates.** Lamb found solutions by obtaining eigenvectors and eigenvalues of the RWA Hamiltonian. Specifically, he sought the three eigenvalues $\varepsilon$ of the determinantal equation

$$\begin{vmatrix} \mathrm{i}\varepsilon & \frac{1}{2}M^* & \frac{1}{2}R^* \\ \frac{1}{2}M & \mathrm{i}\varepsilon + \omega - \omega_{\alpha\beta} & V^* \\ \frac{1}{2}R & V & \mathrm{i}\varepsilon + \omega - \omega_{\alpha e} - \mathrm{i}\gamma \end{vmatrix} = 0, \tag{2.20}$$

and used these to construct solutions to the TDSE, a procedure well known to 19th century mathematicians [Inc56] that subsequently has been termed the use of "dressed states" or "adiabatic states", cf. **Sec.** 2.7.2.

In discussing the dynamics Lamb assumed that initially the states $\alpha$ and $\beta$ held equal populations, and that later, because of radiative decay, state $e$ was devoid of population. He showed a plot of the $\alpha - \beta$ resonance, as a function of magnetic field. It has a shape that later became known as a Fano profile [Fan61], see **Sec.** 2.5.2.

**Trapping.** In discussing his eqn. (219) Lamb showed that when the rf frequency $\omega$ is equal to the Bohr frequency[15] for the $\alpha - \beta$ transition between Zeeman sublevels of the 2S configuration, then the decay rate is zero (i.e. there occurs "population trapping", cf. **Sec.** 2.3.5), and there occurs a "hole" in the spectrum (the *Fano minimum*); cf. also [Fon73]. This superposition can be termed a "dark state", although that terminology was not used by Lamb.

**Summary: Lamb trapped state.** This third Lamb paper, aiming to establish a detailed description not only of the energy structure of hydrogen and its isotopes, set out the basic TDSE of three-state coherent excitation. It made use of what are now termed "dressed states" and identified a superposition state that had the properties subsequently identified as the "dark state": a coherent superposition that undergoes no loss. These structures were not dynamic — there was no consideration of time-varying components, as is essential for STIRAP. Because the collisional interaction $V$ of **Fig.** 2.3$(b)$ is always present, the linkage pattern of the Hamiltonian was a loop rather than a chain.

[15]Note: Lamb referred to "Bohr energy", "Bohr radius", "Bohr magneton" but never "Bohr frequency", only energy differences.

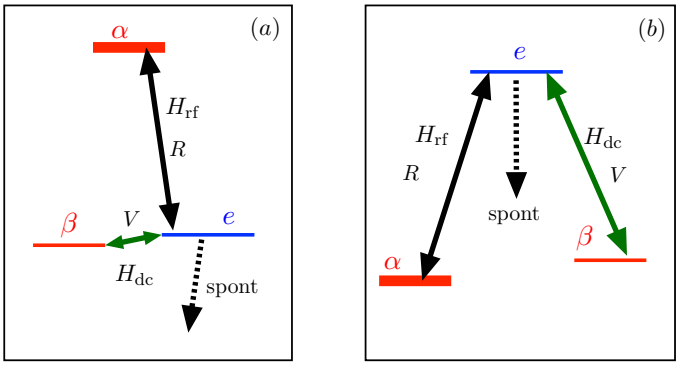


Fig. 2.4. Two presentations of the three-state chain-linkage $\alpha - e - \beta$. $(a)$ With energies ordered as in the experiment. $(b)$ With quantum state locations as needed to give a lambda linkage.

### 2.3.5 The population-trapping state of Lamb

It is instructive to redraw the linkage pattern of **Fig.** 2.3 as shown in **Fig.** 2.4$(b)$. In the rotating-wave picture the vertical positions of the nodes of the linkage pattern (traditionally indicated by horizontal lines) are irrelevant, and so both these frames show linkages of the same system: a three-state chain $\alpha - e - \beta$ in which the middle state $e$ has decay while the terminating states $\alpha, \beta$, are metastable.

The linkage pattern of frame $(b)$ is what has subsequently been termed a lambda linkage, meaning the middle state of the linkage chain lies highest on the diagram. As is now very well established, with the lambda linkage one can introduce an alternative basis of bright and dark states, as in eqns. (1.39), such that all the loss occurs from the bright state $B$ and the dark state $D$ undergoes no spontaneous emission. Lamb, in the penultimate paragraph of his section 72 on p. 274 of [Lam52], states that the decaying state $e$ will remain unpopulated if the two metastable amplitudes satisfy the equation (using here notation of the present article)

$$\tfrac{1}{2} R C_1 + V C_3 = 0. \tag{2.21}$$

This effect, and its key relationship, can be recognized as population trapping (a term that appeared only later): there occurs a particular quantum-state superposition that has no component of the decaying state 2 and is therefore immune to loss or spontaneous emission (i.e. it is dark). Its components depend on the relative magnitudes of two elements of the three-state Hamiltonian.

Notably, the two links of the Hamiltonian, here parametrized by rf field $R$ and static field $V$, would later both originate with monochromatic radiation fields, i.e. fields that have nonzero carrier frequencies that nearly match the Bohr frequencies of the two links, here those of transitions $\alpha - e$ and $e - \beta$. In the work of Lamb (and the LASL group) one of the Bohr frequencies is zero, and the link is by means of a quasi-static electric field. In the first STIRAP work the two fields, $P$ and $S$, were radiation fields, with nonzero carrier frequencies and subsequent STIRAP work has been with either traveling waves or standing waves.

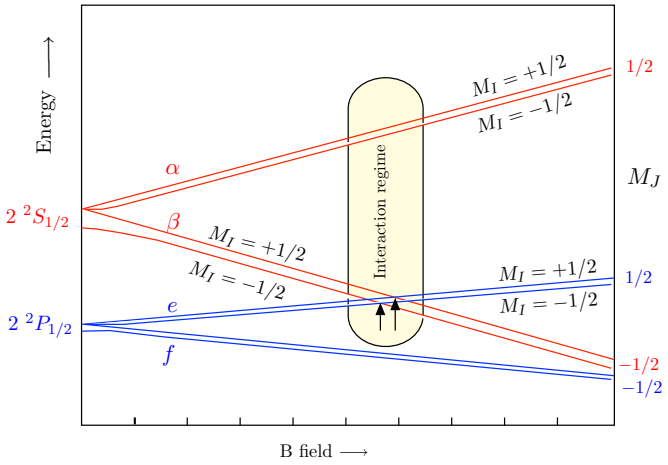


Fig. 2.5. Hyperfine components of 2S and 2P states as a function of magnetic field strength. Yellow cartouche covers the resonance regime of interest. Short vertical arrows mark the locations of resonances in $M_I = -1/2$ and $M_I = +1/2$, used for separating these two states. Redrawn after Fig. 16 of [Lam50]. Reprinted with permission from [Lam50]. Copyright 1950 by the American Physical Society.

### 2.3.6 Hyperfine structure; Nuclear spin filter, 1951, 1967

The interest in population trapping by Lamb and his associates had to do specifically with hyperfine structure and the preparation of spin-polarized particles. The nuclear spin is responsible for a degeneracy that, in the presence of a magnetic field, splits the Zeeman sublevels into hyperfine components. For hydrogen the nuclear spin is $I = 1/2$ and the splitting forms doublets. For deuterium the nuclear spin is $I = 1$ and there occur a triplet of hyperfine components.

**Figure** 2.5 redraws **Fig.** 2.3 to show the hyperfine splitting produced by the nuclear spin. In the regime of interest, as outlined by the yellow cartouche, the nucleus retains a given orientation, unaffected by the radiation. That is, $M_I$ is a "good quantum number", and we need only consider a set of curves with given $M_I$. The system then has the three-state dynamics considered above, in the absence of nuclear spin.

As shown in **Fig.** 2.4 the three states are three Zeeman sublevels, metastable $\alpha$ and $\beta$ and radiatively decaying $e$, in the Lamb notation. Each of these is actually one state of a hyperfine doublet, distinguished by the two values of $M_I$. There are three fields: a static magnetic field that induces Zeeman shifts, thereby making $\beta$ and $e$ degenerate, an rf field that will induce the $\alpha - e$ transition, and a quasistatic velocity-induced electric field that couples $\beta$ to $e$. In the absence of $\beta$ the population in $\alpha$ would rapidly be optically pumped into $e$, from which it would be lost by spontaneous emission. The link between $e$ and $\beta$ will, for appropriate rf frequency (as determined by the Zeeman splitting), prevent the loss from $\alpha$. To use a term introduced a few years later, there occurs a *population trapping state*. This trapping can be made specific to a chosen magnetic spin projection, $M_I$.

### 2.3.7 LASL work, 1967

Starting in 1967 researchers at Los Alamos Scientific Laboratory (LASL) [Ohl67,McK68,Law69, Ohl71] used the properties just mentioned, along with the dynamics discussed in earlier sections above, to devise a procedure in which a specified nuclear spin projection $M_I$ would remain undiminished in an atomic beam as it passed through various fields, while atoms that had other spin projections would undergo "quenching" radiative decay. They use the term "spin filter". None of the papers by Lamb described above use that terminology.

The dynamics of the linkage pattern of **Fig.** 2.3 — that of idealized hydrogen without nuclear spin — was examined in some detail by Ohlsen and McKibben in an unpublished LASL memo of 1967 [Ohl67]. In the simplest case they considered the sublevels (quantum states) $\beta$ and $e$ are degenerate, and the field polarization is such that there is no direct $\alpha - \beta$ linkage, i.e. $M = 0$. Their TDSE was that of Lamb [Lam52], shown above as eqn. (2.18). They provided analytic solutions to these equations subject to the initial condition that all population resides in the quantum state $\alpha$, $C_\alpha(0) = 1$. Like Lamb [Lam52] they expressed these by means of eigenvectors and eigenvalues. They were interested in asymptotic solutions, at time sufficiently long that transients have decayed. They gave the explicit solutions, recognizable as components of the dark state of eqn. (1.21),

$$C_\alpha \to \frac{V}{|R/2|^2 + |V|^2}, \qquad C_\beta \to \frac{-(R/2)}{|R/2|^2 + |V|^2}, \qquad C_e = 0. \tag{2.22}$$

Thus the final disposition of populations depends on the two interaction strengths, one steady rf $(R)$ and one quasi-static $(V)$.

## 2.4 Autler-Townes splitting,1955–78

In 1955 Autler and Townes [Aut55] presented detailed formulas for evaluating the effects of ac fields on quantum states — energy shifts now referred to as the *ac Stark shift* or *dynamic Stark shift*. They pointed out that these shifts would affect spectral lines by splitting the emission or absorption peak into two peaks, now known as an *Autler-Townes doublet* produced by *Autler-Townes (AT) splitting*. This effect, of a doublet produced by a strong interaction probed by a weaker interaction, can be found in eqn. (7.87) of Bloch [Blo55]. When this splitting becomes sufficiently large there occurs a striking minimum of excitation and absorption, subsequently used to advantage in producing electromagnetically induced transparency of a propagating beam, cf. **Sec.** 5 and **App.** F.1 and F.2.

### 2.4.1 The Autler-Townes dressed states

Regarded as an aspect of three-state excitation by two fields, the AT splitting is a characteristic of systems that have two very different Rabi frequencies, such as occurs at the start and finish of a STIRAP pulse sequence or as illustrated for steady fields in **Fig.** 2.14 below (in **Sec.** 2.9.2). It can be observed in any linkage pattern by any technique that probes the energy structure, e.g. photon absorption or fluorescence or electron scattering.

The essentials of the behavior of such a system are readily understood with the aid of eigenstates (dressed states) of the strongly-coupled portion of the Hamiltonian, cf. [Ima89, Sho08,

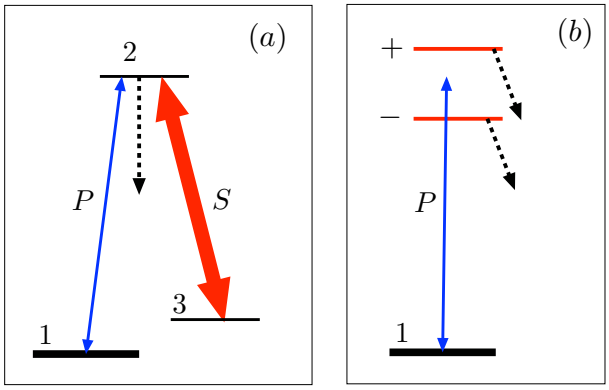


Fig. 2.6. $(a)$ Three-state linkage pattern between bare states 1,2,3. A weak probe field $P$ connects states 1 and 2, a strong field $S$ connects states 2 and 3. (The ordering of energies $E_n$, shown here as a lambda linkage, is immaterial.) $(b)$ The linkages when two dressed states are used for the 2-3 connection. The dashed arrow denotes probability loss, either from spontaneous emission or from photoionization. From Fig. 51 of [Sho08]. Reprinted from [Sho08] with permission.

Sho11]; see **App.** F.2. Figure 2.6$(a)$ shows a three-state linkage pattern, displayed in lambda form, in which a weak probe field $P$ connects a populated ground state 1 with a decaying excited state 2 that, in turn, is linked by a strong field $S$ to a stable third state 3. Using the two AT dressed states $|+\rangle$ and $|-\rangle$ discussed in **App.** F.2 to describe strongly coupled states 2 and 3, we obtain the picture in frame $(b)$, in which the probing $P$ field, resonant with the 1-2 Bohr frequency, is midway between the two lossy dressed states created by the resonant strong $S$ field. At that frequency, $\Delta_P = 0$, and in the limit of very large $S$ field, the $P$ field encounters destructive interference of links to the two dressed-state components that prevent excitation; see **App.** F.2.

### 2.4.2 Optical Autler-Townes splitting, 1978

It was not for another two decades after the 1955 work of Autler and Townes, when lasers were being exploited, that experiments revealed AT splitting at infrared and optical frequencies [Sch75a, Sch75b, Cah76, Piq76, Del78]. **Figure** 2.7 shows an example from a paper by Gray and Stroud in 1978 [Gra78]. Zoller [Zol79] provided detailed theory, illustrated, for noisy lasers, as an example of double optical resonance (DOR) with unequal Rabi frequencies.

The Autler-Townes splitting provides an explanation for the start and end of the STIRAP procedure [Vit01a, Vit01c], when AT splitting produced by the strong field prevents the weak field from causing population changes; see **Sec.** 5.5.

## 2.5 Continuum states, 1958–89

Although STIRAP, like other procedures for coherently manipulating quantum states, deals with just two or three states, a complete set of quantum states must include not only discrete bound states but a continuum in which motional variables are allowed a continuum of kinetic energies.

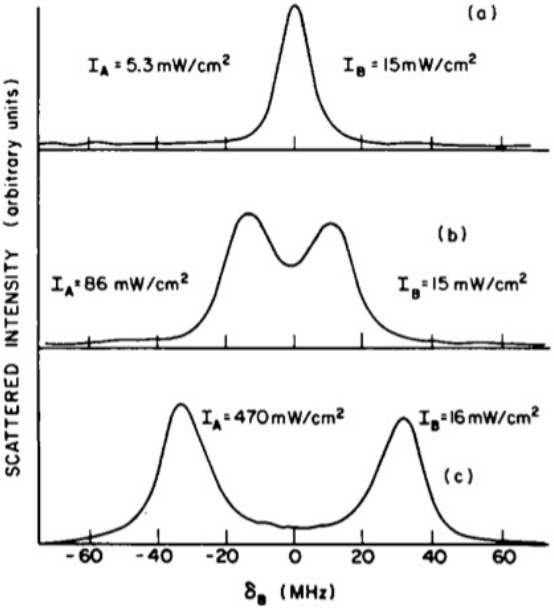


Fig. 2.7. Autler-Townes doublet seen in fluorescence. From Fig. 1 of [Gra78]. Reprinted from [Gra78] with permission from the Optical Society of America. Here the strong $S$ laser is tuned to exact Bohr-frequency resonance, $\Delta_S = 0$, and the detuning $\Delta_P$ (here labeled $\delta_B$) of the weak $P$ field is scanned through its resonance, while measuring fluorescence (i.e. loss) from state 2. As the intensity $I_A$ of the $S$ field increases, from top to bottom frames, the AT splitting increases in direct proportion to $\sqrt{I_A}$, i.e. the magnitude of the $S$-field Rabi frequency. See **Sec.** 5 and 5.2 and **App.** F.2 for discussion of the theory.

The photoionization continuum of an atom and the photodissociation continuum of a molecule are two examples. The continuum of photon directions and energies needed to describe free-space spontaneous emission is another. The inclusion of continuum states, of energy $\varepsilon$, means that the statevector construction of eqn. (1.5) becomes, for a single degree of freedom,

$$\mathbf{\Psi}(t) = \sum_n C_n(t)\, \exp[-\mathrm{i}\zeta_n(t)]\, \boldsymbol{\psi}_n + \int d\varepsilon\, C_\varepsilon(t)\, \exp[-\mathrm{i}\zeta_\varepsilon(t)]\, \boldsymbol{\psi}_\varepsilon. \tag{2.23}$$

With this extension the excited state 2 becomes embedded in a spontaneous-emission (or photoionization) continuum, often treated by means of a probability loss rate $\Gamma_2$, evaluated using "Fermi's Famous Golden Rule Number 2" [Fer50],

$$\Gamma_2 = \frac{2\pi}{\hbar} \varpi |\langle 2|V|\varepsilon_2\rangle|^2, \tag{2.24}$$

where $\varpi$ is the density of states at the resonance energy $\varepsilon_2$.

### 2.5.1 Feshbach theory, 1958–67

The notion of a bound state embedded in kinetic-energy continuum of scattering states has a long history in nuclear physics, where resonances are observed in the elastic scattering of neutrons. A general formulation of scattering resonances of this sort, for both elastic scattering and general reactions (e.g. neutron capture) was presented in a series of papers by Feshbach, starting in 1958 [Fes58, Fes62, Fes67]. His attention was directed to scattering states (for arbitrary projectile) as the initial condition, in contrast to the emphasis on the embedded bound state of Fano (in scattering theory such discrete states are known variously as compound states and collision complexes), but here too the exact eigenstates were constructed.

### 2.5.2 Autoionization; Fano profile, 1961

In 1961 Fano [Fan61] presented a description of autoionization (the spontaneous ejection of a valence electron from an excited state whose energy exceeds the ionization energy) by constructing a quantum state that was a superposition of a discrete atomic excited state and a photoionization continuum, each of which were linked to a stable state by radiative interaction, as shown in **Fig.** 2.8(a). The loop linkage pattern is akin to that of Lamb shown in **Fig.** 2.3: two radiative interactions and a static interaction, but displayed as a letter-V rather than a lambda.

Fano constructed time-independent dressed eigenstates of the Coulomb linkage between bound and continuum states, and evaluated the radiative transition probability into this dressed continuum. The Fano formula for absorption cross section near an autoionizing resonance is [Fan61, Fan65]:

$$\sigma(\epsilon) = \sigma_a \left[ \frac{(q+\epsilon)^2}{1+\epsilon^2} \right] + \sigma_b, \tag{2.25}$$

where $\sigma_a$ and $\sigma_b$ are parameters, $\epsilon$ is a scaled measure of the photon detuning from resonance, $\epsilon = (\omega - \omega_0)/(\Gamma/2)$, and $q$ is the *Fano q parameter.*

The radiative absorption profile for state 1, shown in frame $(b)$, exhibits not the usual Lorentz profile that would be seen with the simple 1-2 linkage, but an asymmetric profile, now termed a *Fano profile*, that exhibits a minimum as well as a maximum as a function of photon frequency. The Fano minimum, an interference effect, can be considered a frequency-dependent dark state.

### 2.5.3 Scattering theory and autoionization, 1967

The Feshbach theory was applied explicitly to photon scattering, as is appropriate to stimulated Raman scattering, autoionization and dielectronic recombination, by Shore [Sho67], who presented formulas for absorption and refractive index, parametrized with $A, B, C, D$, as

$$\sigma(\omega) = C + \frac{(\Gamma/2)B + (\omega - \omega_0)A}{(\omega - \omega_0)^2 + (\Gamma/2)^2}, \tag{2.26}$$

$$n(\omega) - 1 = \frac{Nc}{2\omega} \left[ \frac{(\Gamma/2)A - (\omega - \omega_0)B}{(\omega - \omega_0)^2 + (\Gamma/2)^2} - D \right]. \tag{2.27}$$

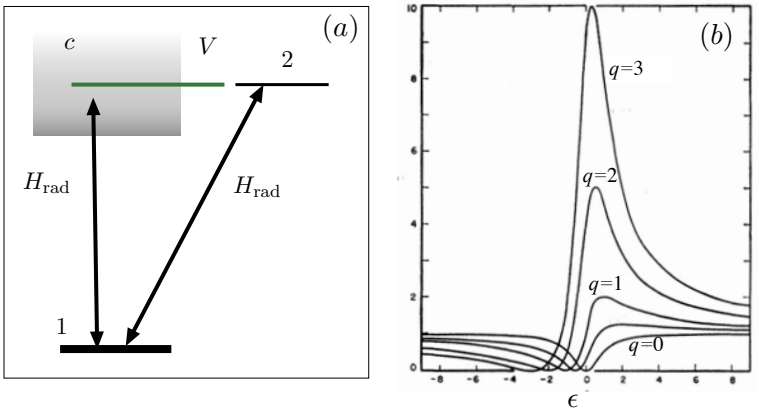


Fig. 2.8. $(a)$ The triangle linkage pattern for autoionization as modeled by Fano [Fan61]: a single field has radiative links to both a discrete state 2 and a continuum, linked by Coulomb interaction $V$ to state 2. $(b)$ The natural spectral-line shape for various values of the Fano parameter $q$. From Fig. 1 of [Fan61]. Reprinted with permission from [Fan61]. Copyright 1961 by the American Physical Society.

The connection between $\sigma(\omega)$ and $n(\omega)$ by means of parameters $A, B, C, D$ is an example of the Kramers-Kronig relationship between real and imaginary parts of causal complex functions. These formulas readily generalize to overlapping resonances [Sho67].

### 2.5.4 Dark resonance, 1973

Fontana and Srivastava in 1973 [Fon73] reconsidered the model used by Lamb [Lam52], namely (with their labeling) two excited states, $a$ and $b$, that are linked to each other by a static interaction $V$. They consider a radiation field that couples state $a$ to ground state $c$ and to a continuum of photon fields. The linkage pattern is shown in **Fig.** 2.9.

The two excited states are regarded as degenerate, and are treated by introducing "perturbed eigenstates" defined as (my notation)

$$|\pm\rangle = [|a\rangle \pm |b\rangle]/\sqrt{2}. \tag{2.28}$$

Using these states and the ground state paired with a photon continuum $|f\rangle$, they express the statevector as

$$\mathbf{\Psi}(t) = C_+(t)|+\rangle + C_-(t)|-\rangle + \sum_f C_f(t)|c\rangle|f\rangle, \tag{2.29}$$

and set up the TDSE. They solve these using Fourier transforms. The solutions introduce a complex-valued radiative decay rate $\gamma$ for state $a$. For very long times the probability amplitude for emitting a photon is

$$C_f(\infty) = H_{fa}\frac{E_f - \frac{1}{2}(E_+ - E_-)}{(E_f - E'_+)(E_f - E'_-)}, \tag{2.30}$$

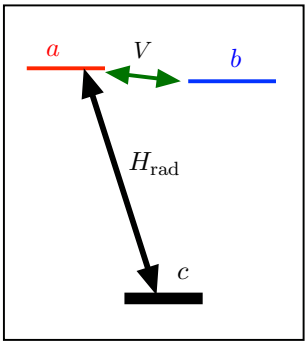


Fig. 2.9. Linkage pattern of ref. [Fon73] for three atomic states. The ground state $c$ is associated with a continuum of photon modes, coupled to excited state $a$ which, in turn, couples by a static interaction $V$ to excited state $b$.

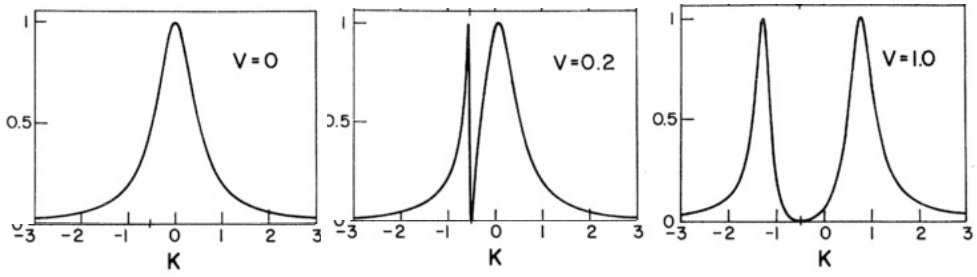


Fig. 2.10. Spectrum of system with linkage pattern of **Fig.** 2.9 and strong-field detuning $\Delta_S$ of -0.5$\gamma$. The plots show, for various values of $V$, the relative probability of photon emission as a function of $K = \omega_P/\gamma$, the photon frequency in units of the decay rate of state 2, here $\gamma$. The emission hole occurs at t he two-photon resonance, $\Delta_P = \Delta_S$. From Fig. 1 of [Fon73]. Reprinted with permission from [Fon73]. Copyright 1973 by the American Physical Society.

where $E'_{\pm}$ is a complex-valued number. They give examples of this spectrum, i.e. the quantity $|C_f(\infty)|^2$, a portion of which is shown in **Fig.** 2.10.

This paper describes a discrete state embedded in a continuum, a model that had been used by Fano in 1961 to describe autoionization [Fan61]. In the Fano application the continuum is that of an ion and a free electron, whereas in the Fontana work [Fon73] the continuum is that of an emitted photon.

One sees here situations whereby photon spontaneous emission is suppressed (a *spectral hole*). This occurs for a particular frequency, and is associated with a particular superposition of excited states. This frequency is not adjustable, although variations of $V$ affect the width of the hole. Thus there are significant differences between what this paper describes and the dark states that appear with optical excitation.

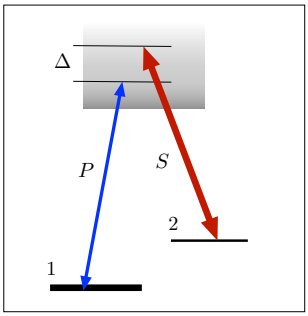


Fig. 2.11. Linkages through a continuum of $P$ and $S$ fields to produce LICS. The carrier frequencies are detuned from the 1-2 two-photon resonance by $\Delta$.

### 2.5.5 Pseudo-autoionization; Laser-induced continuum structure, 1975–82

The embedding of a discrete bound state into an ionization continuum by the Coulomb interaction, as formulated by Fano [Fan61], has a counterpart in the continuum embedding of a discrete state by laster interaction, producing a laser-induced continuum structure (LICS) — a resonance in an otherwise structureless continuum. The possibility of creating a tunable "pseudo-autoionizing" resonance with its enhanced photoionization ionization rate, was discussed by Armstrong, Beers and Feneuille [Arm75], who dealt with quantized fields and photon numbers, and by Heller and Popov [Hel76]. **Figure** 2.11 depicts the process in its simplest form: a strong field $S$ embeds state 2 into an otherwise structureless continuum shared with a weaker probe field $P$ linked to state 1. There is a two-photon transition between states 1 and 2, through the intermediary of the continuum. The two frequencies have a two-photon detuning $\Delta$.

As a result of the discrete state 2 embedded into the continuum, the ionization probability for the probe transition exhibits a Fano profile as a function of the two-photon detuning $\Delta$. The Fano minimum (or "window") in transmission is associated with trapped population (see **Sec.** 2.3). A detailed description of subsequent theoretical work as well as a detailed discussion of the theory appears in a 1990 review by Knight, Lauder and Dalton [Kni90].

Because this is, in part, a coherent process there can occur lossy two-photon Rabi oscillations — the continuum does not entirely eliminate the coherent properties of the two states. An equation for the probability amplitudes for the two states involved given in [Kni90, Pas97, Vit97c] for constant-intensity fields, is

$$\mathrm{i}\frac{d}{dt}\begin{bmatrix} C_1(t) \\ C_2(t) \end{bmatrix} = \tfrac{1}{2}\begin{bmatrix} -\mathrm{i}\Gamma_1 & -\sqrt{\Gamma_1\Gamma_2}(q+\mathrm{i}) \\ -\sqrt{\Gamma_1\Gamma_2}(q+\mathrm{i}) & 2\Delta - \mathrm{i}\Gamma_1 \end{bmatrix}\begin{bmatrix} C_1(t) \\ C_2(t) \end{bmatrix}. \tag{2.31}$$

The parameters $\Gamma_n$ are the single-photon photoionization rates, proportional to radiation intensities. The Fano $q$ parameter is the ratio of the the real to the imaginary part of the two-photon Rabi frequency, complex-valued because it has a contribution from the continuum.

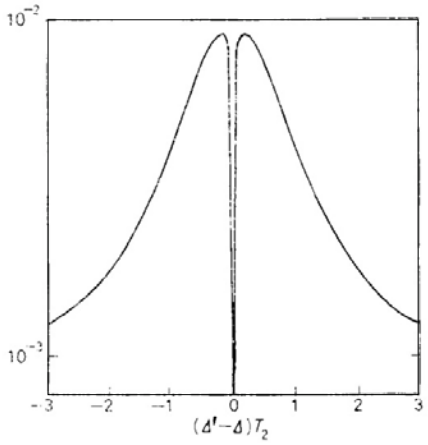


Fig. 2.12. Excited-state occupation as a function of the "resonance parameter" $(\Delta' - \Delta)T_2$, i.e. the two-photon detuning $\Delta_3$ relative to the width parameter $1/T_2$. From Fig. 1 of [Ari76]. Reprinted from [Ari76] with kind permission of Società Italiana di Fisica.

### 2.5.6 Optical dark states, 1976

The experimental recognition of dark states in the three-state lambda linkage, driven by optical fields, is generally credited to work of researchers in Pisa, starting with an observation of dark lines in a pattern of fluorescence from a laser beam passing through sodium vapor [Alz76]. This was followed by theoretical work [Ari76] that, in plotting results from formulas of Brewer and Hahn [Bre75], showed a very distinct dark center (a dark line) within a broad pattern of excited-state excitation as a function of detuning, see **Fig.** 2.12. The dark lines occur where the two-photon resonance condition is satisfied. Their interest was on excitation with fixed Rabi frequencies; they did not consider adiabatic change. This subject was reviewed by Arimondo in 1996 [Ari96]. He referred to dark and bright states as *noncoupled* and *coupled* states, respectively, and used these as basis states for the TDSE.

What makes these three-state optical dark states notably different from two-state superpositions that had been considered earlier is that the three-state system involves two linkages that each originate with a controllable radiation field having nonzero carrier frequency. The linkage pattern is traditionally presented as a lambda whose legs are associated with two Rabi frequencies, each of which derives from a field that has nonzero carrier frequency, i.e. a stimulated Raman scenario.

## 2.6 Further population trapping, 1978–89

Renewed interest in population trapping (but without adiabatic change) followed the growing application of laser technology to the manipulation of quantum states and the Pisa work on dark states. The next subsections refer to notable theoretical developments.

### 2.6.1 Dark and bright states, 1978

The theory of three-state coherent excitation (as contrasted with optical pumping) was discussed by Gray, Whitley and Stroud in 1978 [Gra78]. In treating the TDSE for a three-state lambda linkage they pointed out that, for steady fields, "one linear combination of the ground states is coupled to the excited state by the applied fields, and the other linear combination is decoupled entirely", a clear definition of the bright and dark states of the RWA Hamiltonian. Their eqns. (4) express the dark and bright variables, in the present notation, as

$$C_D(t) = \cos\theta\, C_1(t) - \sin\theta\, C_3(t) \qquad C_B(t) = \sin\theta\, C_1(t) + \cos\theta\, C_3(t), \tag{2.32}$$

with $\tan\theta = \Omega_P/\Omega_S$. Here too the interest was in steady illumination and constant mixing angle $\theta$. They presented figures similar to **Fig.** 2.12.

### 2.6.2 Population trapping, 1982–83

A series of papers by Knight and his colleagues [Col82a, Col82b, Dal82a, Dal82b, Rad82] examined numerous aspects of population trapping (often termed *coherent population trapping* or CPT)[16] in three-state systems driven by two radiation fields. The dark state underlies this trapping. These papers pay particular attention to the effects of fluctuations and decoherence on population trapping. Interest here, and in other contemporary work, centered on steady fields, not temporally offset pulses.

A paper by Morris and Shore in 1983 [Mor83] provided a formalism for determining the presence of one or more dark states in suitable linkage patterns, such as the tripod and the resonant chain. The lambda linkage, with its single dark state, is the simplest example. The Morris-Shore (MS) transformation, when applicable, replaces a more elaborate $N$-state linkage pattern by a set of independent two-state systems; see **App.** E.3.

### 2.6.3 Velocity-selective population trapping, 1988–89

In 1988 Aspect and co-workers [Asp88, Asp89] described and demonstrated a technique for creating a population-trapping superposition of degenerate Zeeman sublevels, labeled $g_-$ and $g_+$, that form the base of a lambda linkage to excited state $e_0$ in which the two interactions originate with circularly polarized light that brings momentum to the atom: the polarization $\sigma_+$ adds $\hbar k$ to the center-of-mass momentum $p$ while the polarization $\sigma_-$ removes an increment $\hbar k$. For a given $z$-direction center-of-mass momentum $p$ three states are linked. Using the notation $|$ atom excitation, cm momentum $\rangle$ the three states are

$$|g_-, p - \hbar k\rangle, \qquad |e_0, p\rangle, \qquad |g_+, p + \hbar k\rangle. \tag{2.33}$$

Figure 2.13 $(a)$ shows the linkage of these states.

As in any lambda linkage, there is a superposition state that is immune to change. When the two fields have equal Rabi frequencies this population-trapping (non-absorbing) state is

$$\mathbf{\Phi}_0(p) = \frac{1}{\sqrt{2}}\left[|g_-, p - \hbar k\rangle - |g_+, p + \hbar k\rangle\right]. \tag{2.34}$$

[16] The acronym CPT is also used for *coherent population transfer*, usually in the context of adiabatic passage.

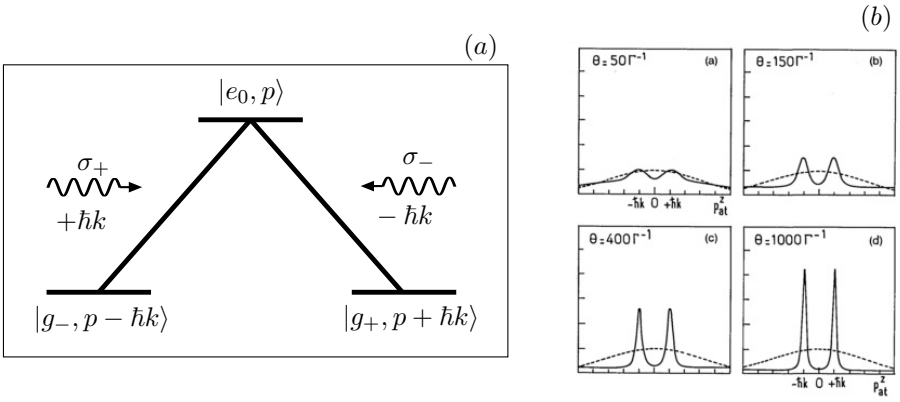


Fig. 2.13. $(a)$ Linkage pattern for the three states involved in velocity trapping, for given center-of-mass momentum $p$. After Fig. 1 of [Asp89] $(b)$ Calculated transverse atomic momentum distribution at a succession of times. The dashed curves show the initial distribution. As the interaction time increases, the height of the double peak at $\hbar k$ (characterizing the cooling process) increases, and its width decreases. From Fig. 5 of [Asp89]. Reprinted with permission from [Asp89]. Copyright 1989, AIP Publishing LLC.

As Aspect et al. point out [Asp89], permanent coherent population trapping, immune to both laser excitation and spontaneous emission, is velocity selective, since it happens only for $p = 0$. For nonzero $p$ there will occur pumping, with a cycle time set by the spontaneous emission rate $\Gamma$, that ultimately terminates in the stable population-trapping state $\mathbf{\Phi}(0)$ of eqn. (2.34), with its components of momentum $\pm\hbar k$. With that change there will occur a narrowing of the velocity distribution — a cooling. Frame $(b)$ shows calculated examples of this cooling for a succession of lifetimes $\Theta = \Gamma t$ [Asp89].

## 2.7 Adiabatic eigenvectors and dressed states, 1970–82

The use of eigenvectors to solve coupled homogeneous ODEs with constant coefficients predates quantum mechanics, and is found in 19th century literature on differential equations where they are known as *characteristic functions*. For application to the mathematics of STIRAP the eigenvectors must accommodate slowly changing ODEs. These adiabatic eigenvectors provide a useful tool for depicting the motion in an abstract vector space.

### 2.7.1 Three-state eigenvectors, 1970–71

Walls [Wal70, Wal71a, Wal71b], treating a quantized-field version of the three-state two-field system, presented formulas for the eigenvectors of the constant RWA Hamiltonian when there is single-photon detuning $\Delta$ but two-photon resonance. With adjustment to the present notation,

his three states are those of eqn. (1.41) and his formulas read

$$\mathbf{\Phi}_0 = [\Omega_S\, \psi_1' - \Omega_P\, \psi_3'] \,/\, \Omega_{\rm rms}, \tag{2.35a}$$

$$\mathbf{\Phi}_\pm = [\Omega_P\, \psi_1' + (\Delta \pm \Upsilon)\, \psi_2' + \Omega_S\, \psi_3'] \,/\, \sqrt{\Omega_{\rm rms}^2 + (\Delta \pm \Upsilon)^2}, \tag{2.35b}$$

where

$$\Omega_{\rm rms} \equiv \sqrt{\Omega_P^2 + \Omega_S^2}, \qquad \Upsilon \equiv \sqrt{\Omega_{\rm rms}^2 + \Delta^2}. \tag{2.36}$$

These formulas give the instantaneous eigenvectors of the slowly varying RWA Hamiltonian. Later articles give alternative expressions for these eigenvectors [Aga79, Rad82, Gau90, Few97]. Walls was interested in the field dynamics and did not comment on the properties of the dark state $\mathbf{\Phi}_0$.

Incidentally, the use of eigenvectors as a technique for solving sets of coupled linear ordinary differential equations with constant coefficients — such as those for multistate excitation by monochromatic light in the RWA — was well established in 19th century literature on differential equations, as discussed by Ince [Inc56].

### 2.7.2 Dressed atoms, 1977

Although others had used the technique for treating three state dynamics [Lam52, Wal70, Wal71a, Wal71b, Whi76], the idea of using eigenstates of the full Hamiltonian (in the rotating-wave approximation) as basis states owes much to the work of Cohen-Tannoudji and Reynard who used such states with density-matrix equations to describe resonance fluorescence and absorption between Zeeman sublevels [Coh77a, Coh77b, Coh92]. They used the term "dressed atom" to describe this basis. When the RWA Hamiltonian becomes time dependent its instantaneous eigenstates become *adiabatic states*, a term used earlier in describing wavefunctions of bound electrons during scattering events. The term "adiabatic state", with a very different meaning, has had a long history in thermodynamics, e.g. [Ran70, Rud94, Jea17].

### 2.7.3 Dressed states, 1979–82

As had Walls in 1970 [Wal70], Agarwal and Jha [Aga79] gave algebraic formulas for the components of the three eigenvectors of the matrix W for the case of two-photon resonance, without noting particular interest in the dark state. Radmore and Knight [Rad82] gave alternative algebraic expressions for these components and permitted two-photon detuning. Fewell et al. [Few97] also presented algebraic expressions for situations in which there is two-photon detuning.

## 2.8 Resonant coherent excitation, 1932–52

One of the ingredients of STIRAP is the availability of detailed descriptions of population changes. Resonant excitation by steady incoherent light was well studied by spectroscopists, as described in the text by Mitchell and Zemansky [Mit34]. The relevant equations were rate equations and the populations were equilibrium values.

### 2.8.1 Majorana-Rabi oscillations, 1932–37

Early in the application of the TDSE came recognition of what are now termed Rabi oscillations [Maj32, Rab37, Sch37]: Formulas for coherently excited populations of a general spin system by a single field were given in 1932 by Majorana [Maj32]. Expressions for population oscillations between the two states of a spin one-half system were given by Schwinger [Sch37] and Rabi [Rab37], who noted these were special cases of the Majorana formula for oscillatory population transfer between arbitrary Zeeman sublevels [Maj32].

### 2.8.2 Double resonance, 1949–52

It was not until the 1950s with the use of microwaves [Bit49] that recognizable coherent-excitation antecedents to the two-field coherent excitation of STIRAP appear, as double-resonance experiments [Bro52]. The availability of lasers accelerated interest in resonant excitation and in manipulating quantum states coherently. Much of that dealt with two-level atoms, as reviewed in the classic monograph of Allen and Eberly [All75].

### 2.8.3 Optical double resonance, 1976

The two-field linkage through a resonant intermediate state as shown in **Fig.** $1.1(c)$, for which eqns. (1.1) and (1.2) hold, is an example of what has been termed *optical double resonance* (ODR), *optical-optical double resonance* (OODR) or *double optical resonance* (DOR), in which two steady (unpulsed) monochromatic optical fields are tuned to resonance with two linked transitions, as discussed extensively [Bro52, Whi76, Zol79].

In 1976 such a system was discussed, for monochromatic fields, by Whitley and Stroud [Whi76], who included various damping and relaxation terms, as treated by a density matrix (their equations were for expectation values of Heisenberg operators, in the RWA). They used eigenvectors of the relevant $9 \times 9$ coefficient matrix, for which they gave some algebraic expressions, and were primarily concerned with steady-state populations and absorption spectra as a function of detunings.

## 2.9 Three-state dynamics, 1970–78

The relevant three-state RWA Hamiltonian and TDSE for steady fields had been presented by Lamb [Lam52], as discussed above. Within a few years there was interest in the operation of three-level masers, and time-averaged populations were evaluated from the three-state TDSE by Javan [Jav57] for a weak probe field in the presence of a strong saturating field, a situation that underlies the Autler-Townes (AT) splitting discussed in **Sec.** 2.4 and the electromagnetically induced transparency of **Sec.** 5 .

### 2.9.1 Coherent excitation; Three-state propagator, 1970–71

Shimoda, in 1970 [Shi70], considered the three-state system linked by two classical monochromatic fields whose frequencies satisfy the two-photon resonance condition. He presented simple

algebraic expressions for the elements of the propagator $\mathsf{U}(t)$, as used to obtain probability amplitudes at arbitrary times after state preparation at $t = 0$:

$$\mathbf{C}(t) = \mathsf{U}(t)\,\mathbf{C}(0). \tag{2.37}$$

The propagator satisfies the TDSE and reduces to the unit matrix initially,

$$\mathrm{i}\frac{d}{dt}\mathsf{U}(t) = \mathsf{W}(t)\mathsf{U}(t), \qquad \mathsf{U}(0) = \mathsf{1}. \tag{2.38}$$

As formulas of Shimoda show, the elements of $\mathsf{U}(t)$ oscillate with a frequency $\Upsilon/2$, where

$$\Upsilon = \sqrt{|\Omega_P|^2 + |\Omega_S|^2 + \Delta_2^2}, \tag{2.39}$$

and so the populations therefore will exhibit oscillations at the *flopping frequency* $\Upsilon$. As with resonant excitation of a two-state system, complete population transfer occurs periodically between the two ends of the chain, with transfer taking place via the intermediate state.

Walls [Wal70,Wal71a,Wal71b], treating the quantized-field version of this coherent-excitation model presented formulas for the oscillatory populations when there is single-photon detuning but two-photon resonance.

### 2.9.2 Population histories, 1976–78

Illustrations of population dynamics for the three-state chain linked by two radiation fields (optical double resonance), with plots of population histories, were first presented by Sargent and Horwitz in 1976 [Sar76] and then by Shore and Ackerhalt the next year [Sho77]. Figure 2.14 shows three examples with unequal Rabi frequencies. The plots, though obtained by numerical solution to ODEs, were what could be obtained by application of the formulas of Shimoda [Shi70]. As with other work of that time, the interest was in steady illumination, with constant and resonant frequencies, not with pulsed excitation. Unequal Rabi frequencies, as occurs with EIT (cf. **Sec.** 5), were considered [Sho77], but only with an eye for population dynamics with steady fields, as illustrated in **Fig.** 2.14. Frame $(a)$ shows the lossless dynamics when the $S$ field is weak, $\Omega_S = 0.3\Omega_P$ and both fields have null detunings, $\Delta_S = \Delta_P = 0$. States 1 and 2 dominate the three-state Rabi oscillations. Frame $(b)$ shows the lossless dynamics when the $P$ field is weak, $\Omega_P = 0.3\Omega_S$, again with null detunings. Here the population remains concentrated in state 1 (partial trapping). Frame $(c)$ shows this same situation but with detuning $\Delta_P = 0.5\Omega_S$, the requirement for resonance with the AT dressed state $|+\rangle$, see **App.** F.2. As the authors said: "Levels 2 and 3 form a tightly coupled pair which oscillate about a time-dependent average population", i.e. there occurs Rabi oscillation between state 1 and a coherent superposition of states 2 and 3, components of the AT dressed state.

**Loss.** The research of paper [Sho77] was motivated by interest in maximizing photoionization yield from the uppermost state of a ladder linkage. The paper showed, with simulation, that tuning the weak field to resonance with what is, in **App.** F.2, termed a dressed AT state would maximize this yield, i.e. maximize population loss. (The paper also showed that there was an optimal loss rate $\Gamma_3$.) **Figure** 2.15 shows snapshots of population loss at a succession of times as a function of the first-step weak-field detuning $\Delta_P$ for the model of frames $(b)$ and $(c)$ of **Fig.** 2.14 but with state-3 loss $\Gamma_3 = 0.1\Omega_S$. The two maxima are components of an AT doublet.

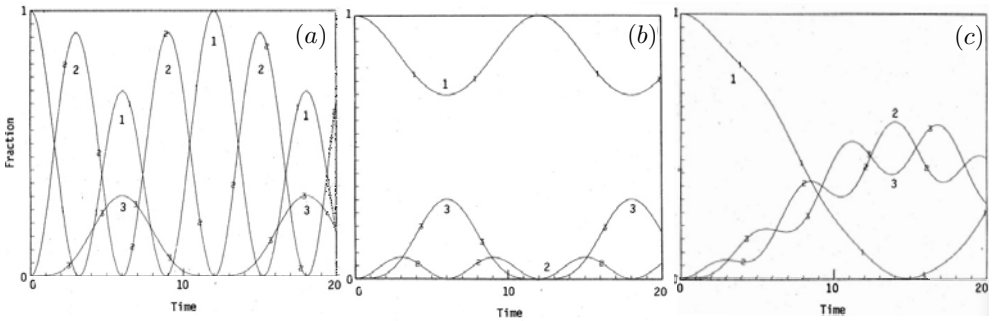


Fig. 2.14. Population histories for three-state system driven by two simultaneous, constant but unequal Rabi frequencies with $\Delta_S = 0$. $(a)$ Weak $S$: $\Omega_S = 0.3\Omega_P$ and $\Delta_P = 0$. Population is predominantly in states 1 and 2. From Fig. 4(a) of [Sho77]. $(b)$ Strong $S$: $\Omega_P = 0.3\Omega_S$ and $\Delta_P = 0$. The population remains largely trapped in state 1. From Fig. 5(a) of [Sho77]. $(c)$ Strong $S$ and retuned $P$: $\Omega_P = 0.3\Omega_S$, $\Delta_P = 0.5\Omega_S$. There is resonant Rabi oscillation between state 1 and a superposition of states 2 and 3 (the AT dressed state $|+\rangle$ of **App.** F.2). From Fig. 7(a) of [Sho77]. (The tiny curve-labels were used with then-available monochrome computer-graphics of 1977 to identify the several curves.) Reprinted with permission from [Sho77]. Copyright 1977 by the American Physical Society.

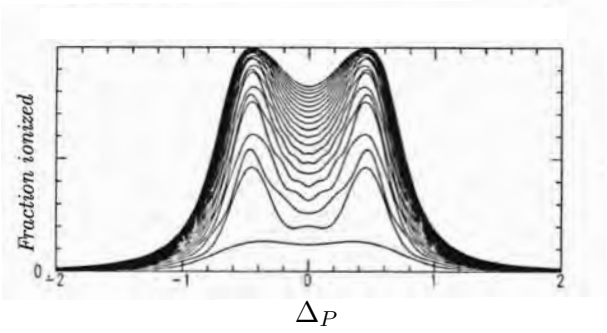


Fig. 2.15. Calculated population loss = $\sum_n P_n(t)$ (fraction ionized) at a succession of times as a function of weak-field detuning $\Delta_P$, for $\Omega_P = 0.3\Omega_S$ and state-3 ionization rate $\Gamma_3 = 0.1\Omega_S$, with $\Delta_S = 0$. The peaks originate with the AT doublet: at those detunings there occur maximum population transfer to and from the lossy uppermost state 3. The loss is less rapid (partial trapping occurs) for $\Delta_P = 0$. Redrawn after Fig. $8(a)$ of [Sho77]. Reprinted with permission from [Sho77]. Copyright 1977 by the American Physical Society. and Fig. 10.3-10 of [Sho90].

## 2.10 Analytic results, 1984–90

Prior to the widespread availability of personal computers, with attendant software for rapidly solving coupled ODEs, theoreticians relied on closed-form algebraic expressions to understand quantum behavior. Such approaches, through analytic approximations for elements of the Hamil-

tonian, still provide useful results, both as a check on numerical calculations and for evaluating limiting cases. The following subsections mention examples. For a review of analytic solutions by Hioe and others to the three-state system see [Hio09].

### 2.10.1 Adiabatic elimination; Two state behavior, 1984

When the detuning term $\Delta$ of eqn. (1.11) is very large the population in state 2 tends to adiabatically follow the populations in states 1 and 2, oscillating rapidly with small amplitude [Sho90]. This makes possible adiabatic elimination of state 2 from the equations, using the approximation obtained by setting to zero the averaged time derivative of that state:

$$\frac{d}{dt}C_2(t) \approx 0, \qquad C_2(t) \approx -\frac{\Omega_P(t)}{2\Delta}C_1(t) - \frac{\Omega_S(t)}{2\Delta}C_3(t). \tag{2.40}$$

The requirement for this approximation is that the magnitude of the detuning be much larger than the Rabi frequencies,

$$|\Delta| \gg |\Omega_P(t)|, \qquad |\Delta| \gg |\Omega_S(t)|. \tag{2.41}$$

The resulting two-state TDSE can be written, with appropriate choice of overall statevector phase, as [Ore84]:

$$\mathrm{i}\frac{d}{dt}\begin{bmatrix} C_1(t) \\ C_3(t) \end{bmatrix} = \tfrac{1}{2}\begin{bmatrix} 0 & \Omega_{\text{eff}}(t) \\ \Omega_{\text{eff}}(t) & 2\Delta_{\text{eff}} \end{bmatrix}\begin{bmatrix} C_1(t) \\ C_3(t) \end{bmatrix}, \tag{2.42}$$

where the effective Rabi frequency is proportional to the product of the two field amplitudes,

$$\Omega_{\text{eff}}(t) = \Omega_P(t)\Omega_S(t)/2\Delta, \tag{2.43}$$

and the effective detuning, time varying, is proportional to the difference of the squares of the two Rabi frequencies (i.e. to the difference of the $P$- and $S$-field intensities)

$$\Delta_{\text{eff}}(t) = [\Omega_P(t)^2 - \Omega_S(t)^2]/4\Delta. \tag{2.44}$$

This intensity-dependent detuning originates with the dynamic Stark shift of states 1 and 3 produced by state 2, not with any sweep of laser frequency, although such a contribution to detuning can readily be incorporated into the equations.

Before the arrival of the pulses both $\Delta_{\text{eff}}(t)$ and $\Omega_{\text{eff}}(t)$ vanish. The effective detuning becomes nonzero when the first pulse arrives, changes sign at the midpoint of the pulse sequence, and remains nonzero until the last pulse ceases. The effective Rabi frequency is pulsed and positive, and is nonzero only when both pulses are present. During its presence the effective detuning undergoes a chirp. **Figure** 2.16 illustrates two examples of population transfer, using both counter-intuitive and intuitive pulse ordering. Because only two states are explicitly involved, not three, these examples do not fit the definition of STIRAP as used in this article.

It is important to recognize that both the two-photon Rabi frequency $\Omega_{\text{eff}}(t)$ and the effective detuning $\Delta_{\text{eff}}(t)$ vary with the fields. When only a single field acts, so that $\mathcal{E}_P(t) = \mathcal{E}_S(t)$, then both these elements of the RWA Hamiltonian are proportional to the field intensity and have a common time dependence, say $f(t)$. Introduction of the increment $d\tau = f(t)dt$ then yields equations with a constant coefficient matrix W [Sho90, Sho11].

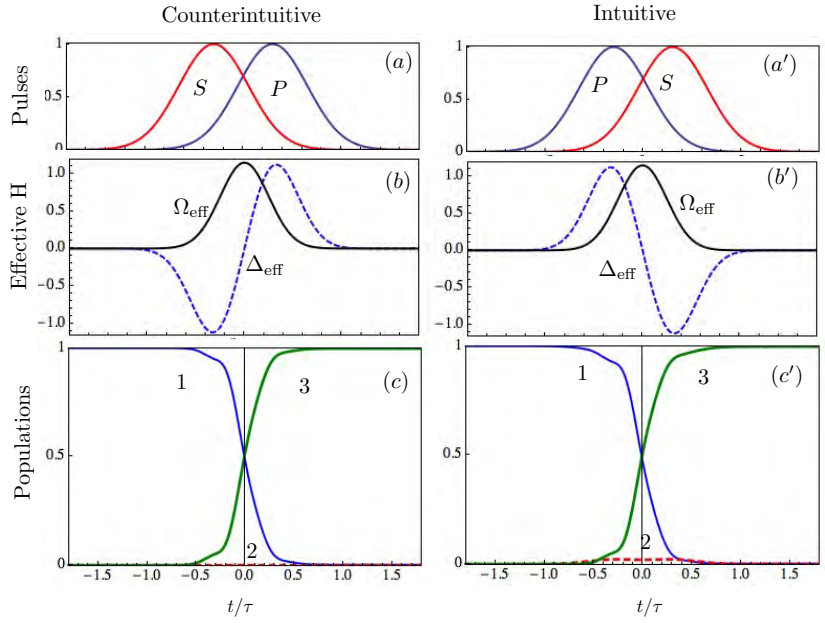


Fig. 2.16. Simulation of three-state dynamics with large single-photon detuning $\Delta$. Left-hand frames are for counter-intuitive pulse ordering, $S$ before $P$. Right-hand frames are for intuitive ordering, $P$ before $S$. Top frames $(a)$,$(a')$ are normalized Rabi frequencies. Middle frames $(b)$, $(b')$ are effective Rabi frequencies and detunings for two-state system obtained by adiabatic elimination. $(c)$, $(c')$ are populations $P_n(t)$ obtained by numerical integration of the three-state equations. If adiabatic elimination were to be exactly valid there would be no population in state 2. Its presence indicates slight failure of the two-state approximation. The two pulses have temporal areas of $60\pi$ and the detuning is $3\times$ the peak Rabi frequencies.

**Adiabatic criteria.** By suitably crafting the $S$ and $P$ fields one could, in principle, produce an effective interaction that acts as a pulsed chirp, thereby moving population adiabatically between states 1 and 3. The probability of this population transfer can be estimated from expressions presented by LZSM [Vit01a, Vit01b],

$$P_3(\infty) = 1 - \exp[-\xi], \qquad \xi \equiv \frac{\pi\Omega_{\rm eff}(t_x)^2}{2|\dot{\Delta}_{\rm eff}(t_x)|}, \tag{2.45}$$

where $t_x$ is the midpoint of the pulse sequence. To have a high probability of population transfer (large $\xi$) it is necessary that the change of effective detuning be very slow,

$$|\dot{\Delta}_{\rm eff}(t_x)| \ll \Omega_{\rm eff}(t_x)^2, \tag{2.46}$$

or

$$2|\Delta| \times |\dot{\Omega}_P(t_x)\Omega_P(t_x) - \dot{\Omega}_S(t_x)\Omega_S(t_x)| \ll [\Omega_P(t_x)\Omega_S(t_x)]^2. \tag{2.47}$$

In the simple case of two identical pulses offset in time the slopes are equal and opposite at $t_x$. An estimate for the terms then gives the inequality

$$2|\Delta| \ll \Omega_0^2 T, \tag{2.48}$$

where $T$ is of the order of the pulse half width. Because $\Delta$ must be much larger than $\Omega_0$ to justify the two-state approximation this inequality requires that $T$ be very long, a condition that poses experimental challenges.

**Relation to STIRAP.** The two-state equations obtained from three-state equations for large single-photon detuning, and their interpretation of chirped adiabatic passage, were noted by Oreg, Hioe and Eberly [Ore84]. Considering the use of temporally offset pulses for this model they conclude that adiabatic inversion could be achieved by appropriate delay between the pulses. Referring to the effective two-state approximation (i.e. large single-photon detunings), they pointed to the need for the two Rabi frequencies to have opposite slopes, and they illustrate this with **Fig.** 2.2$(a)$ appropriate to linearly-sweeping $P$-before-$S$, i.e. "intuitive" ordering of the pulses

Because the chirp direction does not matter in two-state adiabatic passage, the temporal ordering of the pulses is immaterial here. This way of viewing STIRAP with single-photon detuning — mapping it into the well-studied two-state system with controlled time-dependent Rabi frequency and detuning — was discussed by Vitanov and Stenholm [Vit97b]. It gives one connection between STIRAP and the LZ-LZS-LZSM description of adiabatic passage, cf. **Sec.** 2.1.3. But because it only involves two states, and does not depend on pulse ordering, it is not STIRAP as defined here, although it accomplishes complete population transfer with negligible population in state 2. It is *two-photon RAP.*

### 2.10.2 Three-state LZSM model of adiabatic passage, 1986

The classic model of two-state adiabatic passage used by LZSM (see **Sec.** 2.1.3) takes the Rabi frequency $\Omega$ to be constant and the detuning to vary linearly with time, at rate $r$. This model readily extends to three states, as discussed by Carroll and Hioe [Car86a], who took the two fields to have equal Rabi frequencies. Their RWA Hamiltonians, for $N = 2$ and $N = 3$ were

$$\mathsf{W}(t) = \begin{bmatrix} rt & \Omega/2 \\ \Omega/2 & -rt \end{bmatrix}, \qquad \mathsf{W}(t) = \begin{bmatrix} rt & \Omega/\sqrt{8} & 0 \\ \Omega/\sqrt{8} & 0 & \Omega/\sqrt{8} \\ 0 & \Omega/\sqrt{8} & -rt \end{bmatrix}. \tag{2.49}$$

The three-state Hamiltonian describes a situation in which $\Delta_P = \Delta_S = -rt$ in a ladder linkage for which the two fields are both resonant at $t = 0$. **Figure** 2.17 shows the adiabatic and diabatic eigenvalues associated with these matrices[17].

Carroll and Hioe found approximate analytic expressions for the probabilities of the three states at long times, expressed as functions of the "scaling parameter"

$$s = |\Omega|/4\sqrt{r}. \tag{2.50}$$

[17]Note that these formulas for $\mathsf{W}(t)$ require phase choices $\zeta_n(t)$ different from those leading to the $\mathsf{W}(t)$ shown in eqn. (1.11).

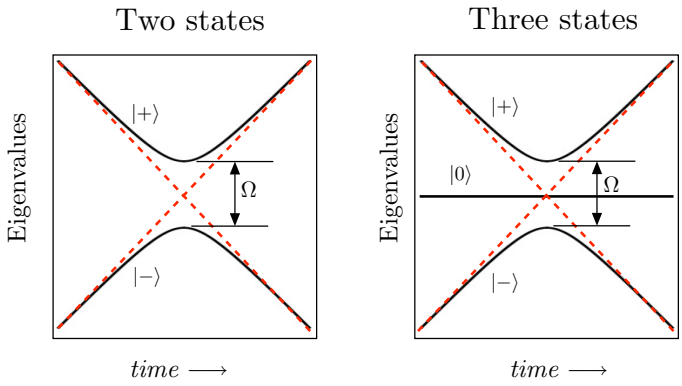


Fig. 2.17. Adiabatic (solid lines) and diabatic (dashed lines) eigenvalues for two-states (left) and three states (right), assuming constant Rabi frequencies and linear chirp, with the RWA Hamiltonians of eqn. (2.49). Redrawn based on Fig. 1 of [Car86a]. Reprinted with permission from [Car86a]. Copyright IOP Publishing. All rights reserved.

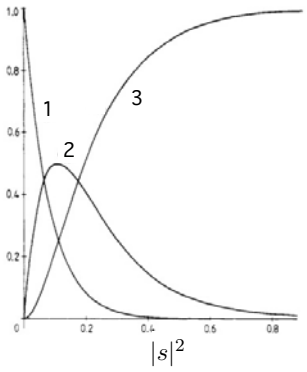


Fig. 2.18. Final populations as a function of the square of the "scaling parameter" $s = |\Omega|/4\sqrt{r}$. Small $s$ produces diabatic passage (the system remains in state 1), large $s$ produces adiabatic passage (the system transitions to state 3). From **Fig.** 2 of [Car86a], with altered curve labels for visibility. Reprinted with permission from [Car86a]. Copyright IOP Publishing. All rights reserved.

**Figure** 2.18 shows their results, for initial population entirely in state 1. As with the two-state system, adiabatic following holds if $|\Omega|$ is large and $r$ is small.

### 2.10.3 Analytic solutions, delayed overlapping pulses, 1984–88

Starting in 1984 Hioe and Eberly [Hio84a], Hioe [Hio84b] and Carroll and Hioe [Car88a], treating coherent excitation of $N$-state systems by pulses having a common analytic time dependence,

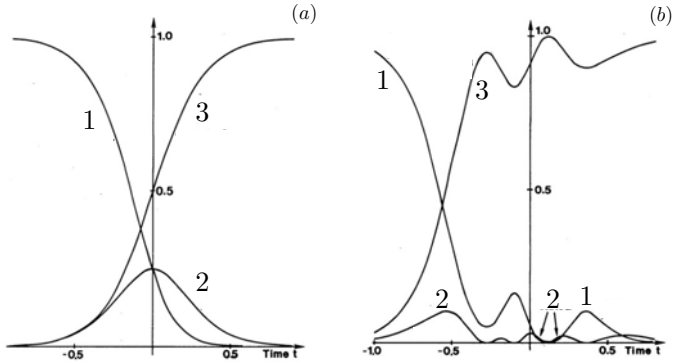


Fig. 2.19. Results of modeling three-state excitation with two offset analytic pulse shapes. From Figs. 2 and 3 of [Car88b], with curve labels 1,2,3 added for clarity. Reprinted with permission from [Car86a]. Copyright IOP Publishing. All rights reserved.

presented exact analytic solutions to the coherent density-matrix equations for a class of $N$-state linkages that included the three-state system with two-photon resonance and swept single-photon detuning. The model is a generalization of the two-state LZSM model of chirped adiabatic passage, cf. **Sec.** 2.1.3, with hyperbolic functions for pulses and detunings, as used by Allen and Eberly [All75]. The review [Hio09] discusses a variety of analytic pulse shapes for which analytic solutions are possible for three-state systems.

Starting in 1986 Carroll and Hioe [Car86c, Car88b] examined analytic solutions to resonant three-state excitation by pulses having different shapes and whose amplitudes were chosen as analytic functions offset in time. The solutions, extensions of those for two-state pi pulses, were expressed in terms of Clausen functions. **Figure** 2.19 shows two examples of results they displayed. Although the evolution of the system can be adiabatic, the presence of chirped detuning sets this apart from STIRAP.

### 2.10.4 Hioe papers, 1981–90

Numerous papers provide supporting formalism for $N$-state adiabatic following, although none fully described what is defined above as STIRAP. These works, many by Hioe, found constants of motion, and dark states, and fitted them into notions of multidimensional Hilbert-space motion by means of a coherence vector [Hio81, Elg82, Hio82a, Hio82b, Hio82c, Hio83a, Hio83b, Hio84a, Hio84b, Ore84]. Notably, several of these papers pointed out the existence of various constants of motion for the density matrix subject to coherent excitation. Indeed, the dark adiabatic state is a consequence of SU(3) group symmetries of the RWA Hamiltonian. Exact expressions for populations subject to offset pulses having analytic forms for envelopes appear in Carroll and Hioe papers [Car88a, Car88b] but the offered solutions have variable detuning and so are not STIRAP.

Although STIRAP fits readily into the formalism of the coherence vector, none of these papers explicitly identified the STIRAP mechanism: a statevector that adiabatically follows a

dark adiabatic eigenvector by means of a pulse sequence in which the $S$ field precedes the $P$ field and the carrier frequencies remain constant, maintaining two-photon resonance. The examples chosen are what have been called RCAP-CHIRAP in **Sec.** 4 or the two-state limit of large single-photon detuning, cf. **Sec.** 2.10.1. However, the paper [Ore84] is prescient for remarking, referring to the effective two-state approximation: "... adiabatic inversion can be obtained simply by designing a proper delay between the pulses" and "... if we wish a practical inversion scheme we must sweep the Rabi frequencies in opposite directions".

Several papers by Hioe and Carroll presented solutions to the three-state system, on two-photon resonance, with analytic expressions for pulse shapes and detuning, but with simultaneous pulses [Hio84b, Car86a, Car86b, Car88a]. Hioe was a co-author on the Kuklinski paper of 1989 [Kuk89] and with Carroll he described in 1990 solutions for delayed pulses, as are needed for STIRAP. For a review of analytic solutions by Hioe and others to the three-state system see [Hio09].

**Multiphoton transitions.** When the single-photon detuning is large (but two-photon resonance applies) the STIRAP process, like other stimulated-Raman processes, can be regarded as a two-photon transition, between two discrete states by way of an intermediate state that receives negligible population — a "virtual level" [Sho79]. More generally a two-photon transition rate involves a superposition of many nonresonant intermediate states [Goe31, Wei33, Gol65], a result traditionally derived using perturbation theory.

The formulas (2.43) and (2.44) are special cases of multiphoton transitions [Wei33, Fai87, Del94] and dynamic shifts [Bac97] that include the effect of all but two discrete quantum states. More generally there occurs a sum over all possible intermediate states and an integral over continuum states. The denominators include both resonances, $\omega_{nm} - \omega$ and anti-resonances, $\omega_{nm} + \omega$. The more general formulas, valid for constant intensity, obtain either from perturbation theory or by approximation to inversions of Laplace transforms [Sho90].

## 3 STIRAP, 1988–90

The development of experimental STIRAP has been described on a number of occasions at seminars by Klaas Bergmann. Here I do not wish to duplicate that history, only to give a synopsis of the three basic initial publications from his research group that first established STIRAP. I have titled the subsection headings to indicate what first appeared in the article, although much other material was discussed.

### 3.1 First experiments, September, 1988

The procedure now known as STIRAP — three-state adiabatic passage in a stimulated Raman process produced by constant-frequency radiation pulses in which the 2-3 interaction $S$ precedes but overlaps the 1-2 interaction $P$ — was first demonstrated in the experimental work of Gaubatz, Rudecki, Becker, Schiemann, Kuelz, and Bergmann published in September, 1988 [Gau88]. **Figure** 3.1 shows the published experimental results, for vibrational excitation of $Na_2$ molecules in a beam. Versions of this figure have appeared subsequently in many articles and reviews [Gau90, Ber98, Vit01a, Vit01b, Sho11], as evidence of the pulse ordering ($S$ before $P$) that distinguishes STIRAP from other population transfer procedures. Numerical modeling agreed with the experimental results that showed the desirability of $S$ pulse preceding, but overlapping, the $P$ pulse. The results were insensitive to small changes of pulse amplitudes, as is characteristic of an adiabatic process.

### 3.2 First theory, December, 1989

The next year a theoretical paper by Kuklinski, Gaubatz, Hioe and Bergmann appearing in December, 1989, [Kuk89], provided a theoretical basis for the experimental results of [Gau88] by using explicit formulas for the elements of the adiabatic dark state, expressed in terms of a mixing angle that ratioed the two pulse amplitudes. These results differed from earlier discussions of the dark state (cf. **Sec.** 2.3) by considering explicit time dependence, and presenting conditions for validity of adiabatic evolution in the three-state linkage, the now-familiar condition that temporal pulse areas should be much larger than $\pi$. They described their results as an example of adiabatic passage and, more importantly, presented the conditions necessary for adiabatic following — the adiabatic criterion that continues to guide experimenters.

Although not mentioned explicitly in [Kuk89, Gau90] the early work showed transfer of the population of an entire set of Zeeman sublevels of a specific rotational level; see **Sec.** 1.3.15.

Further discussion by Carroll and Hioe of analytic pulse shapes and population formulas (involving Bessel functions and hypergeometric functions) appeared in 1990 [Car90]. Writing with submission in February, 1990, they said “A remarkable common result that emerges from all the preceding analytic solutions is that sending two overlapping laser pulses in the counter-intuitive interaction sequence with large pulse amplitudes provides an efficient way for population transfer in a three-state system that is quite independent of the laser-pulse shapes, and that minimizes the occupation probability of the intermediate state 2 . . . ”. They cite the experimental results of Gaubatz et al. in 1988 [Gau88] as an example of this “adiabatic following”.

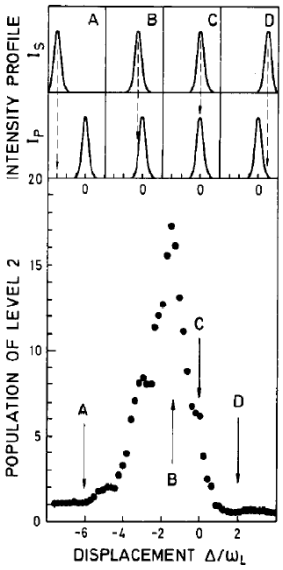


Fig. 3.1. Population transfer as a function of displacement between $S$ and $P$ beams. Frames at the top show the relative placement of the two beams. Transfer is largest when $S$ precedes but overlaps $P$, the so-called counter-intuitive ordering. From Fig. 4 of [Gau88]. Reprinted from [Gau88] with permission from Elsevier. Improved versions appear in [Gau90, Ber98, Vit01a, Vit01b, Sho11].

### 3.3 First acronym, May, 1990

The acronym "STIRAP" first appeared in May, 1990, in an article by Gaubatz, Rudecki, Schiemann and Bergmann [Gau90]. That paper refined the presentation of the sodium-dimer excitation data and developed the theoretical description that has remained the conventional approach to STIRAP, including formulas for the three adiabatic eigenvectors (adiabatic states), which they denoted $|a^-\rangle, |a^0\rangle, |a^+\rangle$. With the present notation (and suppression of explicit time dependence) their formulas for the two additional eigenvectors read (time dependence not shown) supplement eqn. (1.24) [Gau90, Few97]

$$\mathbf{\Phi}_+ = \begin{bmatrix} \sin\theta\,\sin\varphi \\ \cos\,\varphi \\ \cos\theta\,\sin\varphi \end{bmatrix}, \qquad \varepsilon_+ = \tfrac{1}{2}\left[\Delta + \sqrt{\Delta^2 + \Omega_{\rm rms}^2}\right], \tag{3.1a}$$

$$\mathbf{\Phi}_0 = \begin{bmatrix} \cos\theta \\ 0 \\ -\sin\theta \end{bmatrix}, \qquad \varepsilon_0 = 0, \tag{3.1b}$$

$$\mathbf{\Phi}_- = \begin{bmatrix} \sin\theta\,\cos\varphi \\ -\sin\varphi \\ \cos\theta\,\cos\varphi \end{bmatrix}, \qquad \varepsilon_- = \tfrac{1}{2}\left[\Delta - \sqrt{\Delta^2 + \Omega_{\rm rms}^2}\right], \tag{3.1c}$$

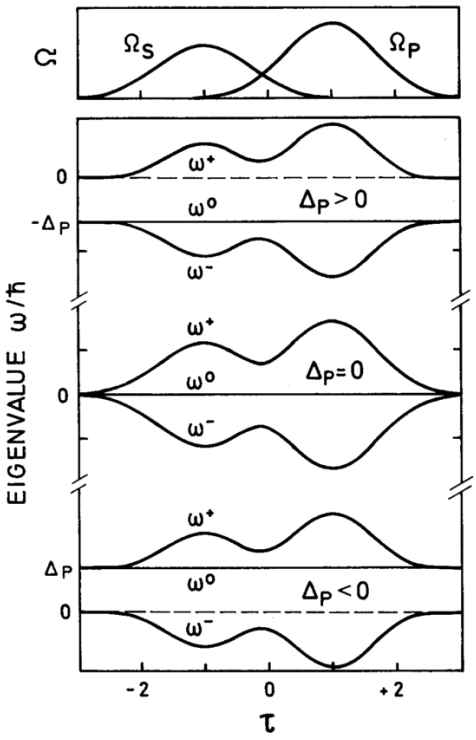


Fig. 3.2. Time dependence of adiabatic eigenvalues, here denoted $\omega^-, \omega^0, \omega^+$, for three choices of the one-photon detuning $\Delta_P$. The eigenvalue $\omega^0$ associated with the dark adiabatic state is constant, but with the phase and energy conventions used for these plots, eqn. (C.4b), it is only zero when $\Delta_P$ is zero. From Fig. 8 of [Gau90]. Reprinted with permission from [Gau90]. Copyright 1990, AIP Publishing LLC.

where

$$\tan\theta = \frac{\Omega_P}{\Omega_S}, \qquad \tan\varphi = \frac{\Omega_{\rm rms}}{\sqrt{\Omega_{\rm rms}^2 + \Delta^2} + \Delta}, \qquad \Omega_{\rm rms} = \sqrt{\Omega_P^2 + \Omega_S^2}. \tag{3.2}$$

They refer to STIRAP as an example of adiabatic passage and adiabatic following [of the statevector $\mathbf{\Psi}(t)$ to the adiabatic eigenvector $\mathbf{\Phi}_0(t) \equiv |a^0\rangle$]. They discussed results of numerical simulation, using a density matrix approach that allow treatment of stochastic effects. **Figure** 3.2 reproduces their Fig. 8 plot of adiabatic eigenvalues, showing the constant eigenvalue $\varepsilon_0(t) = \omega^0$ of the adiabatic dark state. With the phase and energy choices of that paper, eqn. (C.4b), this eigenvalue is equal to the single-photon detuning $\Delta_P$, and is only zero when $\Delta_P = 0$.

Later that year came another use of the acronym, by Hioe [He90], who discussed the relationship of STIRAP to the earlier well-developed *stimulated-emission pumping* (SEP) procedure for producing population transfer [Kit81, Ham86, Bec87].

There followed, within the next few years, tens of papers from the Bergmann group at the University of Kaiserslautern, all treating various aspects of population transfer, or preparation of superposition states, in atomic or molecular beams [Sho91, Cou92, Kuh92, Ore92, Sho92a, Sho92b, Sch93, Mar95, Mar95, Sho95a, Sho95b, Hal96, Mar96, Few97, Una97, Yat97, Gue98b, Hal98, Kuh98, The98, Una98a, Una98b, Vit98a, Vit98b, Yat98a, Yat98b, Hal98]. These works soon extended the concepts of adiabatically evolving dark states to treat multistate linkage chains, angular-momentum degeneracy, and coherent-superposition final states. Soon other groups took an interest in STIRAP, as was noted in early reviews [Ber95, Ber98]. By now the applications of the procedure and further developments of the theory have extended to researchers throughout the world.

## 4 RCAP-CHIRAP: Chirped adiabatic passage. 1983–85

The STIRAP process, as it is defined here, produces adiabatic changes by means of *amplitude* changes of constant-frequency electromagnetic fields. An alternative procedure for accomplishing the robust adiabatic transfer of populations in a three-state chain was proposed by Hioe [Hio83a] and colleagues Oreg and Eberly [Ore84, Ore85] several years prior to the development of STIRAP. Unlike STIRAP, it uses swept detunings, produced by frequency-changing (chirped) radiation pulses, to produce the desired adiabatic changes, as do many other multi-level generalizations of the early two-state rapid adiabatic passage [Vit01a, Vit01b].

Oreg, Hioe and Eberly [Ore84] considered pulses of duration $T$ with common envelopes $\Omega_P(t) = \Omega_S(t)$ and chirped detunings,

$$\Delta_{12}(t) \equiv \Delta_P(t) = a - rt, \qquad \Delta_{23}(t) \equiv \Delta_S(t) = b - rt, \tag{4.1}$$

both resonant at the pulse maxima, $t = T/2$. When $b > a$ the ordering of null detunings (resonances) is 1-2 then 2-3, i.e. that of intuitive population transfer through intermediate state 2. When $a > b$ the order of null detunings, termed "antinatural" or "nonintuitive", is 2-3 then 3-1 and the population transfer proceeds by a two-photon transition with the 1-3 resonance. Referring to the ordering in which LZSM curve-crossings occur when the frequencies are swept they concluded that " ... the 'antinatural' case $a > b$ is the more favorable for adiabatic inversion".

This concept of three-state population transfer using chirped-frequency lasers has subsequently been termed variously *Raman chirped adiabatic passage* (RCAP) [Che95, Che97, Ber98, Vit01b, Lon07a, Tsc07, Kum14] and, particularly for general application to ladder linkages, *chirped-frequency adiabatic passage* (CHIRAP) [Ban94, Dan04].

The RCAP-CHIRAP process has been demonstrated on various molecular excitation situations [Ban94, Dan04]. **Figure** 4.1 shows an example of an experimental demonstration of this technique. As can be seen from that figure (and as was emphasized in earlier theoretical discussion [Sho92a]), the direction of the frequency sweep determines whether the population transfer goes directly between states 1 and 3 or whether it proceeds through the intermediate state 2 or avoids this state (as does STIRAP).

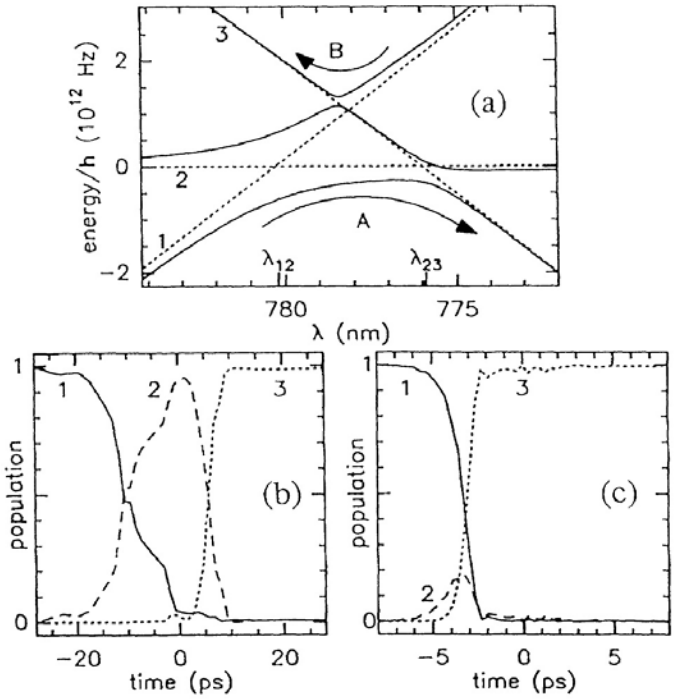


Fig. 4.1. Demonstration of RCAP-CHIRAP. Top frame (a) shows adiabatic eigenvalues (full lines) and diabatic eigenvalues (dotted lines) for three-state ladder linkage with two chirped detunings. Bottom frames show population histories of state 1 (full line), 2 (dashed) and 3 (dotted): (b) for left-to-right detuning sweep and indirect transfer through intermediate state 2. (c) for right-to-left detuning sweep, and direct transfer from 1 to 3. From Fig. 3 of [Bro92]. Reprinted with permission from [Bro92]. Copyright 1992 by the American Physical Society.

## 5 EIT: Transparency, slow light and dark-state polaritons 1989–2000

When one is interested in the effects of matter on the fields as they propagate, it is customary to treat the atoms (or molecules) as a continuous distribution of macroscopic polarization $\mathbf{P}$ and magnetization $\mathbf{M}$ source terms in wave equations derived from Maxwell's equations for electric and magnetic fields. In the absence of free charges and currents the dynamical Maxwell equations read (in SI units with suppression of space and time dependence)

$$\nabla \times \mathbf{E} + \frac{\partial}{\partial t}\mathbf{B} = 0, \tag{5.1a}$$

$$\nabla \times \mathbf{B} - \frac{1}{c^2}\frac{\partial}{\partial t}\mathbf{E} = \mu_0 \nabla \times \mathbf{M} + \frac{\partial}{\partial t}\mu_0 \mathbf{P}. \tag{5.1b}$$

Typically the source terms on the right-hand side of eqn. (5.1b) are treated by introducing constants that characterize the steady-state behavior of aggregates of atoms, often expressed as linear *constitutive relations* [Str41]:

$$\mathbf{P} = \epsilon_0 \chi \mathbf{E}, \qquad \mathbf{M} = \chi_m \mathbf{H}, \tag{5.2}$$

where $\chi$ and $\chi_m$ are electric and magnetic susceptibility tensors — macroscopic averages of single-atom electric and magnetic dipole moments. Such expressions rely on an approximation in which the individual atoms remain in a steady state, i.e. they are not undergoing Rabi oscillations of populations. These relations describe traditional linear optics. When the medium responds transiently to multiple fields they require revision, as discussed in works on nonlinear optics [Arm62, Wal71a, Han79, She84, Boy03].

### 5.1 Pulse propagation and transparency

Within this idealization of steady-state atomic structure the effect on the electric-field envelope of a plane wave of angular frequency $\omega = 2\pi c/\lambda$ traveling along the $z$ axis of a uniform medium is governed, with neglect of higher derivatives, by the equation [Har92]

$$\left[\frac{\partial}{\partial z} + \frac{1}{v_g}\frac{\partial}{\partial t}\right] \mathcal{E}(z,t) = -\mathrm{i}\frac{\pi}{\lambda}\chi\, \mathcal{E}(z,t), \tag{5.3}$$

where the inverse group velocity is

$$\frac{1}{v_g} = \frac{1}{c} + \frac{\pi}{\lambda}\frac{\partial \chi}{\partial \omega}. \tag{5.4}$$

The real part of the (electric) susceptibility $\chi$ alters the group and phase velocities while the imaginary part is responsible for attenuation (or gain). In treating propagation in this way one deals with the atoms as a collective material, characterized by time-independent absorption coefficient and refractive index. The (real) refractive index $\eta$ and absorption coefficient $\alpha$ obtain from the complex-valued $\chi$ for nonmagnetic material through the relationship

$$\sqrt{1+\chi} = \eta + \mathrm{i}\frac{\lambda}{4\pi}\alpha. \tag{5.5}$$

**Transparency.** When the imaginary part of $\chi$ vanishes the material is transparent. This occurs, for example at the Fano minimum of an autoionizing line that has no background absorption.

It can also occur when a second field alters the atomic dipole moments. The three-state chain provides a simple opportunity for such "induced transparency" of the primary field.

The general theoretical description of a three-level lambda linkage with one weak "probe" link was first presented by Weisskopf [Wei33]. It was worked out for general linkages, and susceptibility evaluated, for application to laser amplifiers by Hänsch and Toschek [Hae70] who reviewed earlier work. From steady-state solutions to density-matrix equations they noted that transparency would occur under suitable "hole-burning" conditions described by Bennett [Ben62] and originating with saturation in the strong-field transition, but they also wrote of a "non-linear interference effect ... related to Raman scattering". This is not visible in their illustrations.

Starting in 1990 Harris and co-workers published a series of papers [Har90, Bol91, Har92, Kas95] that described the properties of a three-state chain (experimentally demonstrated with a lambda linkage) in which the two Rabi frequencies were constant and had very different values. (This work modeled a three-state system with loss from state 2 by a TDSE, and used steady-state solutions.) The stronger interaction $S$ (termed in this context the *control* field) acts to create an Autler-Townes doublet, while the weaker interaction $P$, connected to the initially populated state, probes this doublet. The effect is visible as transparency for the probe field $P$, induced by the strong field $S$. (Atoms starting in state 2 undergo gain, and the overall effect for the three-state system is *lasing without inversion*, LWI [Ima89, Scu89, Koc92, Har97].)

Extensive reviews exist for this *electromagnetically induced transparency* (EIT), most commonly observed in propagation but also seen as hindered excitation [Har97,Mar98,Luk03,Fle05].

Although three-state chains with constant unequal Rabi frequencies had been discussed earlier [Sho77, Sho90], the interest had been in population histories rather than the susceptibility (i.e. coherences) and field propagation that has been identified with EIT.

### 5.2 Absorption, population trapping and Fano profile, 1989–92

Imamoglu and Harris [Ima89], modeling the lambda linkage with loss from state 2 (and stable states 1 and 3), showed that the absorption of a weak $P$ field in the presence of a strong $S$ field had a Fano profile as a function of $P$-field detuning. The Fano minimum, at two-photon resonance, represented complete transparency, the result of destructive interference between excitation paths from state 1 to the two dressed states of the 2-3 link and thence into the loss continuum. (Vitanov et al. [Vit01a] show that when both states 2 and 3 are lossy the interference minimum is no longer zero, and that in fact there will be enhanced absorption when $\Gamma_3 \gg \Gamma_2$.)

In 1992 Lounis and Cohen-Tannoudji [Lou92] treated population trapping in the lambda linkage using scattering theory, for the situation when one of the two steady fields is much weaker than the other, as occurs with the Autler-Townes effect and EIT. They showed that the weak-field scattering amplitude is the sum of two amplitudes, identifiable with Rayleigh scattering and Raman scattering. The interference between these two scattering amplitudes produces a Fano profile in the total scattering cross-section.

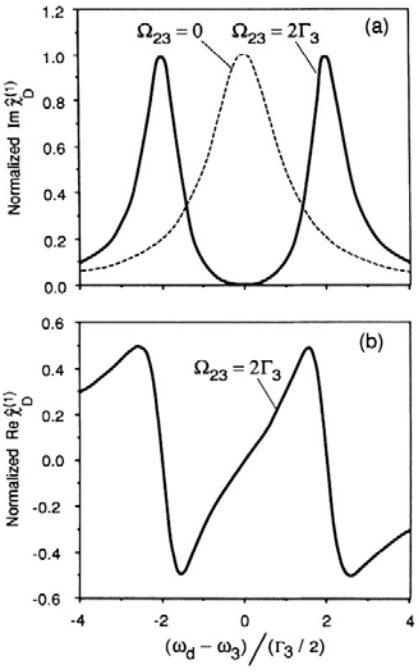


Fig. 5.1. Real (a) and imaginary (b) parts of the susceptibility for a weak probe beam in the presence of a strong control field. The dashed line of frame (a) shows the absorption in the absence of the control field. When that strong field is present there is a transparency window. Normalization is to the peak value of the imaginary part of the susceptibility. From Fig. 2 of [Har90] Reprinted with permission from [Har90]. Copyright 1990 by the American Physical Society. and Fig. 5 of [Har97]. Reproduced with permission from [Har97]. Copyright 1997, American Institute of Physics.

### 5.3 Slow light, 1999

The elimination of absorption, parametrized by the imaginary part of a complex-valued susceptibility, accompanies a reduction of the group velocity, controlled by the real part of the susceptibility. The result is *slow light* [Hau99, Fle05, Khu10]. This phenomena has explanation in models that treat the atomic structure as steady, responding to the thermal environment and external fields, but not undergoing coherent excitation such as Rabi oscillations. **Figure** 5.1, from [Har90], shows the absorption and associated dispersion for a weak probe passing through material in which there is strong Autler-Townes splitting. At the center of the frame, where the AT splitting gives maximum transparency (null absorption), there is a large positive derivative of the susceptibility, and hence an appreciable decrease in the group velocity.

### 5.4 Stored light: Dark state polaritons, 2000

The slowing of a light pulse results from alteration of the polarization field $\mathbf{P}(\mathbf{r},t)$ (a macroscopic distribution of induced dipole moments) that serves as a source for the electric field in the Maxwell equations describing wave propagation, (5.1). When one deals with radiation pulses that are shorter than the relaxation times that obscure coherences of atomic excitation it is necessary to treat both the effect of fields on atoms and of atoms on fields. This is done, for example, with coupled Maxwell-Schrödinger or Maxwell-Bloch equations [All75, Sho90]. The polarization field must be evaluated from the time-varying single-atom dipole moments, customarily regarded as uniformly distributed with a number density $\mathcal{N}(\mathbf{r})$. For the electric dipole moments this is[18]:

$$\mathbf{P}(\mathbf{r},t) = \mathcal{N}(\mathbf{r})\mathrm{Re}\,\langle\mathbf{d}(\mathbf{r},t)\rangle = \mathcal{N}(\mathbf{r})\mathrm{Re}\,\sum_{nm}\mathbf{d}_{nm}C_n(\mathbf{r},t)C_m(\mathbf{r},t)^*\exp[\mathrm{i}\zeta_m - \mathrm{i}\zeta_m]. \quad (5.6)$$

This field has frequencies specified by the phases $\zeta_n$ as modulated by the probability amplitudes. The field amplitudes that are part of the Rabi frequencies must be evaluated subject to alteration by the induced dipole moments that constitute the $\mathbf{P}$ field.

For many purposes it suffices to treat the $P$ field as a pulsed traveling plane wave, steadily exposed to a strong and steady control field $S$ in a Raman linkage. The description of the weak electromagnetic $P$-field coupled to the distribution of polarization is then obtained by introducing a new field, a coherent superposition of electromagnetic (photonic) variable, parametrized by a spatially and temporally varying Rabi frequency $\Omega_P(z,t)$, and an induced dipole moment, proportional to a two-state atomic coherence (an off-diagonal element of the density matrix), $C_3(z,t)C_1(z,t)^*$ and known, for historic reasons, as a "spin wave". The superposition is termed a *dark-state polariton* [Fle00, Vit01a, Fle02],

$$F(z,t) = \cos\Theta(z,t)\Omega_P(z,t) - \sin\Theta(z,t)\sqrt{\alpha c\Gamma}\,C_3(z,t)C_1(z,t)^*. \quad (5.7)$$

Here $\alpha$ is the absorption coefficient, $\Gamma$ is the incoherent relaxation rate and the mixing angle is

$$\Theta(z,t) = \frac{\sqrt{\alpha c\Gamma}}{\Omega_S(z,t)}. \quad (5.8)$$

The dark-state polariton obeys the propagation equation [Vit01a]

$$\left[\frac{\partial}{\partial t} + c\cos^2\Theta(z,t)\frac{\partial}{\partial z}\right]F(z,t) = 0. \quad (5.9)$$

When $\Omega_S(z,t)$ is approximately uniform in $z$ this equation describes the propagation of a quasiparticle with velocity

$$v(t) = c\cos^2\Theta(t). \quad (5.10)$$

When a polariton propagates in an EIT medium, it preserves its amplitude and shape, but its properties can be modified by simply changing the intensity of the control field $S$. By adiabatically

[18] To include effects of different environments the right-hand side must include a sum over these, as in eqn. (1.43).

rotating $\Theta$ from 0 (a strong $S$ field) to $\pi/2$ (a weak $S$ field) an experimenter causes an initially purely electromagnetic field ($F = \Omega_P$) to become a pure polarization field ($F = \sqrt{\alpha c \Gamma} C_3 C_1^*$), and the propagation velocity to drop from the vacuum speed $c$ to zero. Thus by slowly altering the control field an experimenter can diminish the group velocity and, in the limit, bring the polariton to rest, thereby capturing the initial probe field within the medium — it is then a purely atomic excitation in which the initial photonic state has been mapped onto a spatially distributed atomic two-state excitation (a spin wave). As long as the trapping procedure is sufficiently smooth (i.e. adiabatic) the process is lossless and coherent. The mechanism is readily recognized as a variant of the STIRAP process [Fle99].

Unlike the conventional absorption process, this coherent procedure retains all of the phase information from the initial pulse. The recording of the field is reversible, reproducing the initial field, thereby providing a means of storing and retrieving images that include full phase information of the field [Jul04, Bei10, Hei10, Hed10, Gro11].

### 5.5 AT, EIT and STIRAP stages

At the start of the STIRAP process the $S$ field is much stronger than the $P$ field, conditions that produce an Autler-Townes splitting of the $P$ transition. This AT effect, and the concomitant EIT (see **App.** F.1) can be seen in various ways, by means of any technique that probes the energies of the three states, e.g the fluorescence (scattering) experiments of Gray and Stroud [Gra78] that produced **Fig.** 2.7. One such probe is the EIT produced on the $P$ field as it propagates. Because of the AT splitting and the consequent EIT transparency the weak $P$ field is not absorbed and, in turn, induces no population transfer; see **App.** F.2.

Similar reasoning applies to the late stage of STIRAP, when the $S$ field is negligible (or zero): there occurs an AT splitting of the $S$ transition, an EIT null of excitation, caused by the strong $P$ field, which prevents the $S$ field from altering populations. This is manifested, amongst other ways, in EIT of the (weak) $S$ field.

Between these two extremes any description of the dynamics must treat the two fields together, as does picturing adiabatic following of the dark adiabatic state.

Not only does EIT provide an intuitive picture of STIRAP, but as Fleischhauer pointed out [Fle99], the procedure of STIRAP is needed to prepare the most complete spatial distribution of dark states needed to produce the best EIT.

### 5.6 Simultons, matched pulses and adiabatons, 1981–95

When the pulsed $P$-field becomes comparable in strength to the $S$ field the two must be treated jointly, along with the atoms of the media. The use of three-state systems as media for propagation of two collinear beams of coherent radiation has an extensive history. In 1981 Konopnicki and Eberly [Kon81] discussed solutions to three-level Maxwell-Bloch equations that have the character of simultaneous different-wavelength shape-preserving pulses (optical solitons), which they termed *simultons*: two pulses of identical shape that propagate without loss or change. They described analytic pulse shapes involving sech and tanh functions applied to resonant Cook-Shore pulses [Coo79], and supplemented these with extensive numerical results. For simulton propagation to occur, both the pulses and the medium have to be suitably prepared.

In 1992 Harris [Har93] pointed out that population-trapped atoms can cause two optical pulses, of arbitrary shape, to acquire identical envelopes after a characteristic propagation distance. He termed these paired fields *matched pulses*.

In 1994 Hioe and Grobe [Hio94] presented exact analytic solutions for a pair of resonant shape-preserving solitary-waves that can propagate stably without loss through a medium of three-state atoms having lambda or letter-V linkages or through five-state systems having letter-M or letter-W linkages. Their results, based on Jacobi elliptical functions with tanh and sech as special cases, provided a great variety of pulse pairs, which they termed matched-solitary-wave pairs (MSP). The corresponding atomic evolution ranged from complete depletion to complete population return.

Later in 1994 Grobe, Hioe and Eberly [Gro94] presented further analytic forms of resonant two-pulse excitation in three-state systems. The pulse shapes, based on sech and tanh, and termed *adiabatons*, rely on maintenance of the two-photon adiabatic condition based on the three-level trapping state, eqn. (G.22). Initiated with a long-lasting $S$ field and a later $P$ pulse the paired complementary pules travel with a common group velocity for long distances limited by nona-diabaticity. In 1995 Eberly [Ebe95] provided further discussion, noting that adiabaticity allowed population-trapping states of atoms to excite population-trapping fields states.

Harris commented on some of these works in connection with his review of EIT [Har97].

## 6 Contemporary STIRAP

The various elements mentioned in **Sec.** 1.3 as essential characteristics of STIRAP do not give an adequate picture of the technique as it is used at present. This section provides, not an overview of all applications, which is not feasible, but a sense of how original limitations have been extended, while still maintaining the essence of the process. It therefore gives a partial picture of what the term STIRAP means at this time.

### 6.1 Partial STIRAP

The goal of complete population transfer has, for some applications, shifted to creation of a specified superposition of states 1 and 3 by partially completing the STIRAP population transfer, a process termed *fractional-STIRAP* (F-STIRAP) [Vit99,Dre09]. A conceptually simple example is available with the linkage pattern shown in **Fig.** $1.10(a)$. In this example the pulses turn off at the same rate. Alternatively the two pulses can be forced to terminate abruptly at a moment when the mixing angle $\theta(t)$ has a desired value.

If one is interested not so much in population transfer but in maximizing the induced dipole moment at the two-photon frequency $\omega_{13}$, as created by the coherence $\rho_{13}(t) = C_1(t)C_3(t)^*$, then the optimum pulse sequence will be "half-STIRAP", with $\theta(T) = \pi/4$.

The $S$ and $P$ pulses can also be obtained from a single pulse whose polarization changes from right- to left-polarization; see **Fig.** 1.10.

### 6.2 Reverse STIRAP

The original three-state STIRAP proceeded by aligning the statevector with the dark adiabatic eigenvector $\mathbf{\Phi}_0(t)$, as will happen when the $S$ field precedes the $P$ field and population initially resides in state 1. When there is nonzero single photon detuning $\Delta$ it is possible to induce complete population transfer with pulses that have the opposite ordering, $P$ preceding $S$ (the intuitive pulse ordering). **Figure** 6.1 shows an example, from the original STIRAP paper of Gaubatz et al. [Gau90].

As pointed out by Vitanov and Stenholm [Vit97b], having mind coherent excitation in which spontaneous emission is negligible, "in the adiabatic limit both the intuitive and counter-intuitive pulse orders produce complete population transfer for nonzero intermediate-level detuning." Relevant formulas for the eigenvectors $|\pm\rangle$, for Rabi frequencies much smaller than the detuning magnitude as happens at the start and end of the pulse sequence, depend on the sign of the detuning [19] . When the detuning is positive, $\Delta > 0$, then $\varphi \to 0$ and the formulas read

$$\mathbf{\Phi}_+(t) = \begin{bmatrix} 0 \\ 1 \\ 0 \end{bmatrix}, \qquad \varepsilon_+(t) = +|\Delta|, \tag{6.1a}$$

$$\mathbf{\Phi}_-(t) = \begin{bmatrix} \sin\theta(t) \\ 0 \\ \cos\theta(t) \end{bmatrix}, \qquad \varepsilon_-(t) = 0. \tag{6.1b}$$

[19] Expressions for the eigenvalues and the angle $\varphi$ when $|\Delta| \gg \Omega_{\rm rms}$ require evaluation of $\Delta + |\Delta|$.

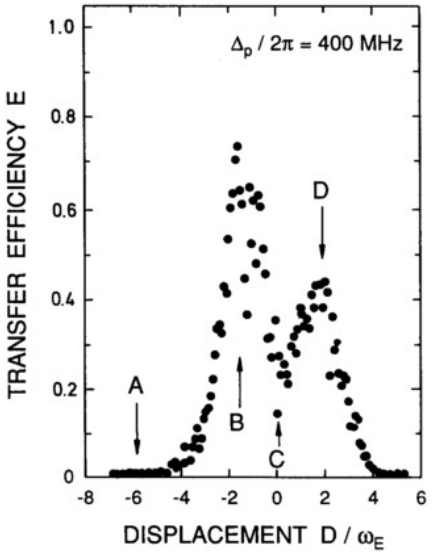


Fig. 6.1. Transfer efficiency (relative excitation probability) vs displacement between plus centers (pulse delay) when there is large single-photon detuning, but two-photon resonance. Letters A and B marks $S$-before-$P$ pulse sequences (B is detuned STIRAP), letter C marks simultaneous pulses and letter D marks $P$-before-$S$ pulse ordering. From Fig. 5 of [Gau90]. Reprinted with permission from [Gau90]. Copyright 1990, AIP Publishing LLC.

Initially and finally the degenerate null eigenvalues of $|0\rangle$ and $|+\rangle$ identify these as being linked with degenerate states 1 and 3; the linkage depends on the mixing angle $\theta(t)$. When detuning is negative, $\Delta < 0$, then $\varphi \to \pi/2$ and the formulas read

$$\mathbf{\Phi}_+(t) = \begin{bmatrix} \sin\theta(t) \\ 0 \\ \cos\theta(t) \end{bmatrix}, \qquad \varepsilon_+(t) = 0, \tag{6.2a}$$

$$\mathbf{\Phi}_-(t) = \begin{bmatrix} 0 \\ -1 \\ 0 \end{bmatrix}, \qquad \varepsilon_-(t) = -|\Delta|. \tag{6.2b}$$

Now it is the degenerate null eigenvalues of $|0\rangle$ and $|-\rangle$ that occur with states 1 and 3.

By controlling the mixing angle $\theta(t)$ an experimenter can move population adiabatically $1 \to 3$ with either $S$ before $P$ or $P$ before $S$ [Gau90]. However, the history of state-2 population differs in the two sequence, as one can appreciate by regarding the eigenvector construction for

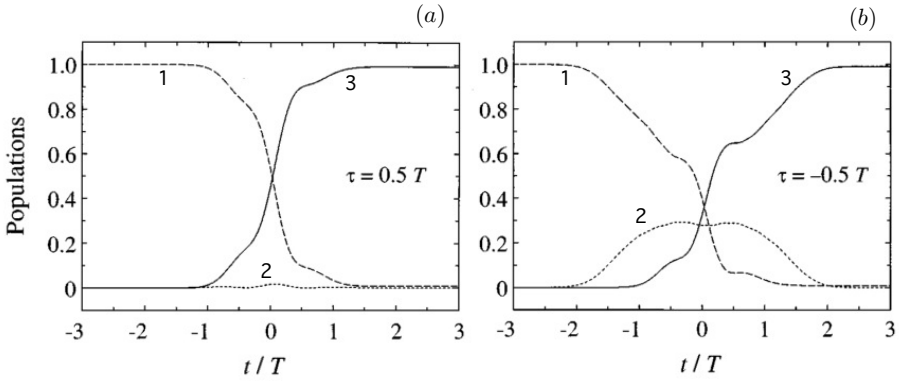


Fig. 6.2. Simulations of population transfer in three-state chain, with nonzero single-photon detuning $\Delta$, produced by adiabatic following with offset pulses $(a)$ Counter-intuitive ordering, $S$ before $P$. $(b)$ Intuitive ordering, $P$ before $S$. From Fig 3 of [Vit97b]. Reprinted with permission from [Vit97b]. Copyright 1997 by the American Physical Society.

resonance or when $|\Delta| \ll \Omega_{\rm rms}$ so that $\varphi \to \pi/4$,

$$\mathbf{\Phi}_+(t) \approx \frac{1}{\Omega_{\rm rms}(t)\sqrt{2}} \begin{bmatrix} \Omega_P(t) \\ \sqrt{2} \\ \Omega_S(t) \end{bmatrix}, \qquad \varepsilon_+(t) \approx \tfrac{1}{2}\Omega_{\rm rms}(t) \tag{6.3a}$$

$$\mathbf{\Phi}_-(t) \approx \frac{1}{\Omega_{\rm rms}(t)\sqrt{2}} \begin{bmatrix} \Omega_P(t) \\ -\sqrt{2} \\ \Omega_S(t) \end{bmatrix}, \qquad \varepsilon_-(t) \approx -\tfrac{1}{2}\Omega_{\rm rms}(t) \tag{6.3b}$$

Both of these eignvectors have "bright" components of state 2, and this will contribute to the population during a pulse sequence.

The possibility of producing complete population transfer using either pulse ordering allows an experimenter to reverse the population transfer — to return population from state 3 to state 1 with this same $S-P$ pulse sequence, thereby restoring the initial state, apart from a phase. In this situation the first pulse sequence is counter-intuitive and the second is intuitive. For this reversing of population transfer the initial statevector alignment is with a bright adiabatic eigenvector, i.e. one that has a component of state 2. The process, termed *bright-state STIRAP*, *backward-STIRAP* or *B-STIRAP* [Vit97b, Kle08, Gri09, Bor10, Sca11a, Sca11b, Sho11] requires one-photon detuning and places transient population into the intermediate state. **Figure** 6.2 shows examples of population histories with nonzero detaining $\Delta$, using either counter-intuitive ordering, $(a)$ or intuitive ordering, $(b)$.

The population histories resulting from B-STIRAP are like those obtained from the chirped detuning of the RCAP-CHIRAP process. **Figure** 6.3 shows, in the right-hand frame, an example of the population histories for an example of B-STIRAP [Bor10]. The appearance is hardly distinguishable from the example of CHIRAP shown in the left-hand frame [Ore84], but the Hamiltonian is different: in B-STIRAP the detuning remains fixed while in this CHIRAP exam-

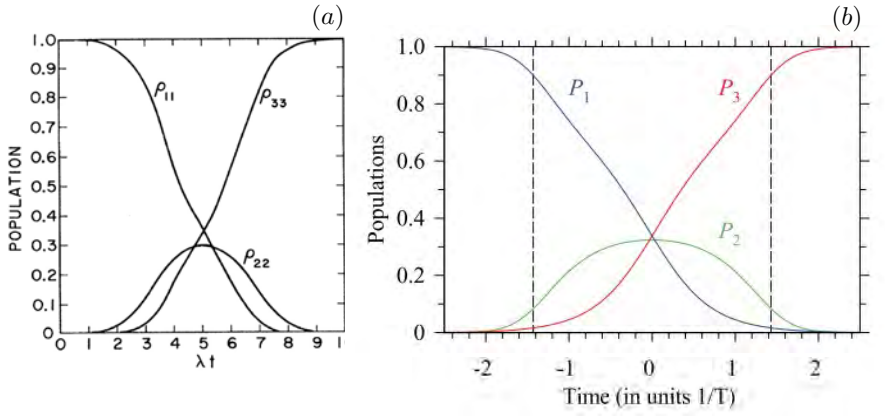


Fig. 6.3. $(a)$: Simulation of RCAP-CHIRAP. From Fig. 1 of [Hio83a], Reprinted from [Hio83a] with permission from Elsevier. And Fig. 4 of [Ore84]. Reprinted with permission from [Ore84]. Copyright 1984 by the American Physical Society. $(b)$ Simulation of bright-state (or backward) STIRAP. From Fig. 2 of [Bor10]. Reprinted with permission from [Bor10]. Copyright 2010 by the American Physical Society.

ple the detuning varies linearly.

### 6.3 Inducing particle structure change

The original experiments defining STIRAP by population transfer between vibrational states in a molecular beam [Gau90] were soon extended to electronic transitions in beams of neutral atoms [Rub91, Mar96] and then to trapped ions [Mau04, Sor06, Wun07] and to color centers in solids [Got06, Kle07, Ale08]. STIRAP has also became a tool for manipulating quantum states of quantum dots [Hoh00] and superconducting nanostructures [Kis04, Pas04, Sie06, Sil09, Sie09, Fal13]. Recently STIRAP has been demonstrated using X-ray pulses to excite isomeric states of atomic nuclei [Lia11].

### 6.4 Additional states and interactions

The original three-state two-field model has been extended in several ways to include additional states and interactions, while retaining characteristics of STIRAP. The basic three-state model of STIRAP readily extends to systems that have Zeeman sublevels [Sho95a, Mar95, Mar96]. Redistribution of populations amongst these sublevels produces coherences [Par93, Una98a, Law98, Vew03, Kis05, Vew07a, Vew07b]. Rice and coworkers [Kob98, Kur01a] considered more elaborate multistate linkage patterns.

**Transfer state.** A condition for successful population transfer with STIRAP is that the statevector adiabatically follow an instantaneous eigenstate of the RWA Hamiltonian. More generally, for application to $N$-state systems, there must exist an *adiabatic transfer state* that connects adiabatically the initial state to the target final state [Mar96, Vit98a].

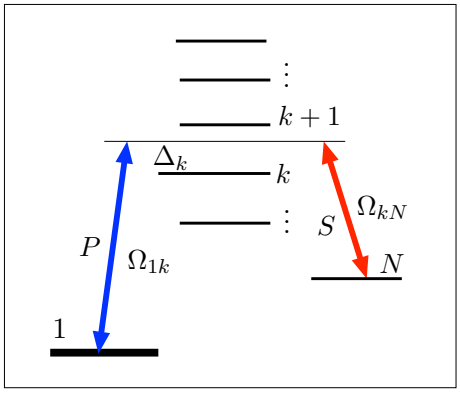


Fig. 6.4. $N$-state linkage pattern for multiple intermediate states.

**Multiple intermediate states.** The idealization of a single intermediate state is not always well satisfied, and very soon after the first work on STIRAP Coulston and Bergmann [Cou92] considered the effect of two intermediate states. This was extended by Vitanov and Stenholm [Vit97a] who treated a set of intermediate states forming a superposition of multiple lambda linkages, each between the same initial and final states, but with different intermediate states and hence with different detunings $\Delta_k$, $P$-field Rabi frequencies $\Omega_{1k}$ and $S$-field Rabi frequencies $\Omega_{kN}$, as shown in **Fig.** 6.4. The TDSE for the probability amplitudes reads

$$\mathrm{i}\frac{d}{dt}C_1(t) = \sum_k \tfrac{1}{2}\Omega_{1k}(t)C_\mu(t), \tag{6.4a}$$

$$\mathrm{i}\frac{d}{dt}C_\mu(t) = \Delta_k C_\mu(t) + \tfrac{1}{2}\Omega_{1k}(t)C_1(t) + \tfrac{1}{2}\Omega_{kN}(t)C_N(t), \tag{6.4b}$$

$$\mathrm{i}\frac{d}{dt}C_N(t) = \sum_k \tfrac{1}{2}\Omega_{kN}C_\mu(t). \tag{6.4c}$$

Vitanov and Stenholm [Vit97a] showed that when the mixing-angle ratio $\Omega_{kN}(t)/\Omega_{1k}(t)$ is independent of $k$ there exists a null-eigenvalue adiabatic state and the possibility of STIRAP. Even when this constraint on the ratio is not fulfilled it is possible, under appropriate conditions, to have successful adiabatic population transfer.

Numerous authors have discussed the generalization from a few discrete states to a quasi-continuum of states [Car92] and to a true continuum, as described by eqn. (2.31) [Nak94, Pas97, Vit97c, Yat97, Una98b, Una00b, Pet05, Pet07].

**Longer chains.** Following the initial development of STIRAP subsequent work has extended the notion of multistate adiabatic passage to longer chains, still with two sequentially-pulsed radiation fields that act on alternating links of the chain [Sho91, Ore92, Vit98a, Vit98b]. The single dark state that makes possible the adiabatic following of STIRAP is also found in $N$-state chains with any odd-integer $N$ [Sho91]. A simple example is the five-state letter-M (or double

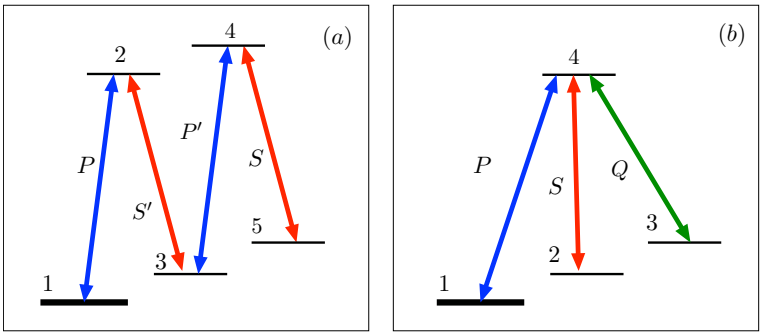


Fig. 6.5. $(a)$ Five-state *letter-M* linkage (or *double lambda*) with two fields (one dark state). $(b)$ Four-state tripod linkage with three fields and linkages only to state 3 (two dark states) .

lambda) linkage shown in **Fig.** 6.5$(a)$. The single dark state, superposing states 1, 3, 5, ..., has no component of states 2, 4, .... The use of two fields, with $S$ before $P$, will produce complete population transfer between state 1 and state $N$, as in STIRAP.

A variant on this chain system places the $P$ pulse only on the 1-2 transition and the $S$ pulse only on the $N$ to $N-1$ transition, and has a "straddling" pulse (or pulses) that connects in chainwise fashion states 2,3, ..., $N-1$. The primed linkages of **Fig.** 6.5$(a)$ are examples of the straddling pules, to extend in duration beyond the $S$ and $P$ pulses. It is possible with this linkage to produce transfer from state 1 to state $N$ by what has been termed straddle-STIRAP [Mal97]. The mechanism for successful adiabatic population transfer involves $N-2$ dressed states that superpose all but states 1 and $N$ and that serve as intermediate states for a set of $N-2$ simultaneous three-state lambda linkages [Vit98b].

**Tripod and fan linkages.** An $N$-state *fan linkage* (also known as an "N-pod", extending the nam "tripod"), in which $N-1$ states all have links only to a single state by means of $N-1$ interactions, has $N-2$ dark states. An example is the five-state tripod linkage of **Fig.** 6.5$(b)$. This system has two degenerate dark states, without any component of state 4. Adiabatic following with a generalization of STIRAP will produce a controllable superposition of states 1,2,3 [Una98a, Una99].

## 6.5 Spatial changes

The first experiments using STIRAP induced population transfers between vibrational levels of diatomic molecules. More complicated molecules often exist in two physical forms, enantiomers that are mirror images of each other and whose stable configurations — each with vibrational states — are separated by a potential barrier. Ohta and colleagues [Oht02] have considered using STIRAP to move population between the two enantiomers, thereby altering the chirality of the molecule. **Figure** 6.6 illustrates the system, an example of the linkage pattern of **Fig.** 1.10$(a)$.

One can extend the use of vibrational wells to a variety of systems, including optical lattices in which single atoms or ions are held and undergo vibration. The coupling between potential

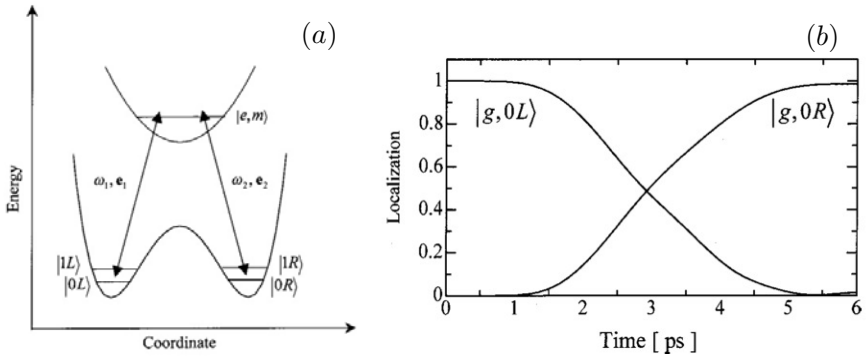


Fig. 6.6. Example of enantiomer transfer via STIRAP. The connection between vibrational states of two enantiomers depends upon the polarization of the field: the $S$ and $P$ transitions have opposite circular polarizations. A single laser field with smoothly altering polarization produces the desired STIRAP. $(a)$ The linkage pattern produced by two polarizations. From Fig. 1 of [Oht02]. $(b)$ Simulation of the population change induced by a STIRAP pulse sequence obtained by varying polarization of a constant amplitude field. From Fig. 5c of [Oht02]. Reprinted with permission from [Oht02]. Copyright 2002, AIP Publishing LLC.

wells, via tunneling, depends sensitively upon their separation, which can be adjusted by an experimenter. The simple example of three such potential wells allows a STIRAP procedure for population transfer between well 1 and well 3, bypassing the intermediate well. **Figure** 6.7 shows an example, from Eckert et al. [Eck04]. In such situations there is no carrier frequency and no RWA; the basic TDSE receives its time dependences from manipulation of trapping-potential dimensions.

## 6.6 Altering the fields

All of those experiments were concerned with the probability amplitudes describing internal structures of particles — the three quantum states that, together with two fields, underly the STIRAP mechanism. But other experimenters have examined the changes to the *fields* that accompany these changes in quantum states. The earliest of these experiments extended the static phenomena of electromagnetically induced transparency (see **Sec.** 5) in which a strong control field ($S$) alters the propagation characteristics (group velocity and absorption) of a weak probe field ($P$), to produce *slow light* [Hau99,Fle05,Khu10]. By controlling the $S$ field an experimenter controls a matter-field superposition, a *dark-state polariton*, in the host matter [Fle00,Fle02]. See **Sec.** 5.4.

STIRAP has been used to alter particular frequency components of the macroscopic polarization field $\mathbf{P}(\mathbf{r}, t)$ in transparent material, thereby enhancing such nonlinear effects as frequency generation [Lon07b, Suc08, Tse10a, Tse10b, Por12].

Like the original Raman scattering process, STIRAP transfers a photon from the $P$ field to the $S$ field. When the weak field is the standing-wave field of a cavity, the strong field can alter the photon structure of the cavity field in a controlled way — making nonclassical field

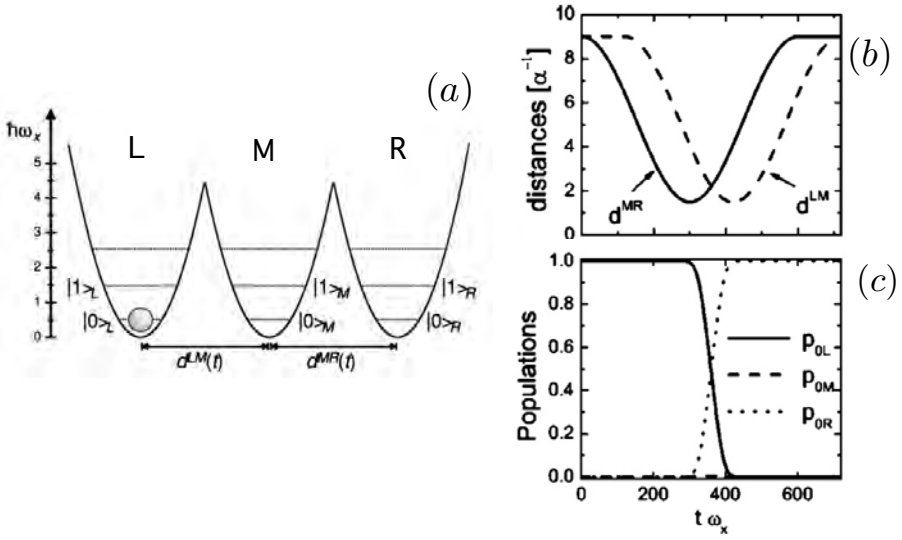


Fig. 6.7. Example of transfer between localizing potentials in which separation-dependent tunneling serves as the interaction. $(a)$ The three wells, L, M and R, with energies of vibrational states $|0\rangle$ and $|1\rangle$. Coupling is by tunneling, adjusted by varying separations $d^{LM}$ and $d^{MR}$. $(b)$ Simulated time dependence of separations for STIRAP. $(c)$ Simulated population histories showing transfer from L to R, avoiding M. From Figs. 1 and 2 of [Eck04]. Reprinted with permission from [Eck04]. Copyright 2004 by the American Physical Society.

states [Sha01,Lar05,Lin08,Toy11,Nog12] or making photons on demand [Kuh02,Mau04,Bar09, Mat09, Vas10].

## 6.7 Waveguides and fibers

A waveguide for electromagnetic radiation is a structure that allows free propagation along one axis, taken to be $z$, while confining the field in the transverse $x, y$ plane. An optical fiber is one example. With assumptions of slow variation of the material properties along the $z$ direction, as expressed by the refractive index $n(x, y, z)$ of the fiber cladding and $n_0$ of the core, a monochromatic electric field amplitude $\mathcal{E}(x, y, z)$ having free-space wavelength $\lambda$ obeys a paraxial wave equation. With the introduction of a scalar amplitude $\Psi(x, y, z) = \exp[-\mathrm{i}k_0 z]\mathcal{E}(x, y, z)$, where $k_0 = 2\pi n_0/\lambda$, the relevant equation is [Kaw01, Vor03],

$$\mathrm{i}\hbar_e \frac{\partial}{\partial z}\Psi(x, y, z) = \left[-\frac{\hbar_e^2}{2}(\nabla_\perp)^2 + V(x, y, z)\right]\Psi(x, y, z). \tag{6.5}$$

Here $\hbar_e = \lambda/(2\pi n_0) = 1/k_0$ is a length scale, $(\nabla_\perp)^2$ is the transverse Laplacian, and

$$V(x, y, z) = \frac{1}{2}\left[1 - \left(\frac{n(x, y, z)}{n_0}\right)^2\right]. \tag{6.6}$$

This equation is analogous to the TDSE but with distance $z$ as the independent variable rather than time $t$. In the transverse direction the field-confinement expressed by the dimensionless $V(x, y, z)$ is analogous to a spatially varying potential energy. It allows evanescent fields, and thereby quantifies coupling between two adjacent waveguides. The counterpart of the basis state expansion (1.5) of the statevector that underlies the derivation of the coupled equations (1.10) from the TDSE is the expansion of an electric field amplitude in terms of spatial field modes $\psi_n(x, y)$,

$$\Psi(x, y, z) = \sum_n c_n(z) \exp[-\mathrm{i}\zeta_n(x, y, z)]\, \psi_n(x, y), \tag{6.7}$$

leading to a set of coupled-mode ordinary differential equations [Yar73,Hau91]. For three modes, coupled to nearest neighbors in a planar layout and with suitable idealization of the tunneling action, the equations can be presented as

$$\mathrm{i}\frac{d}{dz}\begin{bmatrix} c_1(z) \\ c_2(z) \\ c_3(z) \end{bmatrix} = \tfrac{1}{2}\begin{bmatrix} 0 & \Omega_P(z) & 0 \\ \Omega_P(z)^* & 2\Delta_2(z) & \Omega_S(z) \\ 0 & \Omega_S(z)^* & 2\Delta_3(z) \end{bmatrix}\begin{bmatrix} c_1(z) \\ c_2(z) \\ c_3(z) \end{bmatrix}. \tag{6.8}$$

The controllable spatial variation of the elements $\Omega_n(z)$ and $\Delta_n(z)$ originate with the tunneling between adjacent waveguides; they are spatial analogs of the Rabi frequencies and detunings of the TDSE. There have been experimental demonstrations of both STIRAP [for which $\Delta_3(z) = 0$ and $\Delta_2$ is constant] [Lon06a, Lon06b] and RCAP-CHIRAP [Lon07a] as well as results analogous to more general changes of multi-state systems [Del08, Cir12, Cir13]. **Figure** 6.8 shows an example of STIRAP with optical fibers [Lon07a, Lon09].

### 6.8 The defining equations

The usefulness of the original STIRAP concepts to other situations — to various interactions including classical optical fields — suggests that a useful and less restrictive definition of STIRAP can be made just from the set of three coupled ordinary differential equations (1.9), (1.10) or (6.8), together with initial conditions and timing conditions on the coefficient matrix $\mathsf{W}(t)$ [the Rabi frequencies of eqn. (1.11)]. The manipulation of tunneling interactions of optical trapping potentials offer an example, cf. **Sec.** 6.5.

The optical fiber equations serve as another example of this view, in which propagation distance $z$ replaces time $t$ as the independent variable and tunneling replaces electromagnetic interactions but the properties of the system otherwise map from the STIRAP equations, see **Sec.** 6.7.

**Torque and STIRAP.** Traditional torque equations, (1.30), deal with three real variables treated as a vector $\mathbf{r}$ and three controlling parameters, treated as a vector $\boldsymbol{\Upsilon}$. The equations of resonant STIRAP are of this form when one component of the torque vector $\boldsymbol{\Upsilon}$ is zero — traditionally component $\Upsilon_2$; see **Sec.** 1.3.5. Therefore any three variables $r_1, r_2, r_3$ subject to such an equation of motion are capable of mimicking fully resonant STIRAP. It is only necessary that the initial values be taken such that $\boldsymbol{\Upsilon} \times \mathbf{r} = 0$ and that the subsequent variation of $\boldsymbol{\Upsilon}$ be slow.

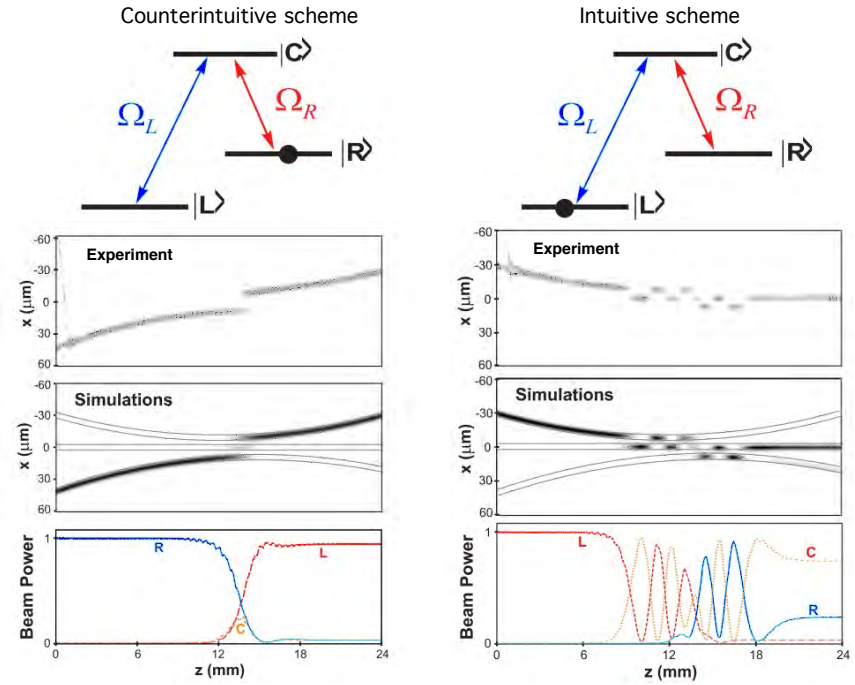


Fig. 6.8. Field transfer in optical fibers. Left hand frames illustrate STIRAP. Right hand frames illustrate Rabi oscillations from intuitive ordering of interactions. From Fig. 4 of [Lon07a] Reprinted with permission from [Lon07a]. Copyright 2007 by the American Physical Society. and Fig. 8 of [Lon09]. Reprinted from [Lon09] with permission of John Wiley and Sons.

**Two-state STIRAP.** Still another example is the mapping of two-state excitation into STIRAP equations, through the three Bloch equations [Lai96, Vit97b, Vit06, Den12]. The three real-valued Bloch variables $u, v, w$, defined in terms of two-state probability amplitudes $c_n$ as [Fey57]

$$u + \mathrm{i}v = 2c_1 c_2^*, \qquad w = c_2 c_2^* - c_1 c_1^*, \tag{6.9}$$

satisfy the three coupled equations (cf. **Sec.** 2.2.2)

$$\frac{d}{dt}\begin{bmatrix} u \\ v \\ w \end{bmatrix} = \begin{bmatrix} 0 & -\Delta & 0 \\ \Delta & 0 & -\Omega \\ 0 & \Omega & 0 \end{bmatrix}\begin{bmatrix} u \\ v \\ w \end{bmatrix}. \tag{6.10}$$

By comparison, the probability amplitudes $C_n$ for fully resonant three-state excitation obey s three-state TDSE that can be written as (note the reordering of variables)

$$\frac{d}{dt}\begin{bmatrix} -C_3 \\ -\mathrm{i}C_2 \\ C_1 \end{bmatrix} = \frac{1}{2}\begin{bmatrix} 0 & -\Omega_S & 0 \\ \Omega_S & 0 & -\Omega_P \\ 0 & \Omega_P & 0 \end{bmatrix}\begin{bmatrix} -C_3 \\ -\mathrm{i}C_2 \\ C_1 \end{bmatrix}. \tag{6.11}$$

Thus when we make the identification of variables $C_n$ as

$$C_3 - C_2 = -(u + \mathrm{i}v) = -2c_1c_2^*, \qquad C_1 = w = c_2c_2^* - c_1c_1^*, \tag{6.12}$$

and the identification of the two Rabi frequencies as

$$\Omega_S = 2\Delta, \qquad \Omega_P = 2\Omega, \tag{6.13}$$

we find that the two-state system obeys the resonant equations of a three-state system — those for which we can initiate a STIRAP process. To do this we begin with the initial two-state conditions

$$w(-\infty) = -1, \quad u(-\infty) = v(-\infty) = 0, \tag{6.14}$$

and apply a pulsed detuning $\Delta$ ahead of an overlapping pulsed Rabi frequency $\Omega(t)$. The result of STIRAP will be a coherent superposition of the two states, as expressed by the variable $u(t)$:

$$u(+\infty) = -1. \tag{6.15}$$

As noted in reviews [Vit01a, Vit01b], the dark-state adiabatic following that is characteristic of the early STIRAP has subsequently been extended to systems that comprise longer chains than three, and for which the linkage pattern is more complicated than an unbranched chain. Here too the acronym STIRAP has often been used. Although the equations deal with two quantum states, they explicitly involve three variables and are identical with the STIRAP equations.

## 7 Conclusion

The benefits of adiabatic passage — relative insensitivity to details of pulse shape, temporal area and timing — make STIRAP attractive as a means of quantum-state manipulation. Raman processes proceed through a lambda linkage, with spontaneous-emission loss from state 2, and so too did the original STIRAP implementation. However, the ladder linkage uses the same equations and timings: it serves equally well for robust adiabatic transfer, justifying the labeling as STIRAP. Both population transfer and coherence preparation benefit from the adiabatic pulse sequences that mark STIRAP. With single-photon detuning present combinations of adiabatic and diabatic following can produce robust quantum changes; not all of these fit the definition of STIRAP used here.

Application of the STIRAP procedure to systems other than discrete states of atoms and molecules require only that system changes be described by the three ODEs of eqns. (1.10 ) and (1.11 ) with $t$ some independent variable, and that the parameters $\Omega_P$ and $\Omega_S$ be suitably sequenced and slowly varying.

The STIRAP mechanism relies on a number of concepts whose history predates the announced demonstration, theoretical description and naming of the first STIRAP [Gau88, Kuk89, Gau90]. Each of these predecessors to STIRAP has an extensive literature and can, in retrospect, be seen as having one or more attributes of STIRAP, but individually they are not the same as STIRAP because they lack some part of the defining characteristics:

- A three-state chain of linked quantum states (or other variables).
- Two pulsed interactions (e.g. constant-carrier radiation fields) offset in time (or other variable) but overlapping.
- Constant detunings [diagonal elements of $\mathsf{W}(t)$].
- Maintained two-photon resonance [or equivalent diagonal elements of $\mathsf{W}(t)$].
- Adiabatic following.
- A populated dark or population-trapping eigenvector of $\mathsf{W}$; minimal population in state 2.

The 1952 paper of Lamb [Lam52] presented the basic physics of population trapping in a three-state chain. It differs from later works in having one of the two field carrier frequencies be zero, i.e. one field is quasi-static while the other is radiative and monochromatic, rf or optical. It does not matter whether the dark-state pairs are excited states or metastable states or some combination, because in the RWA all that matters is the linkages, not the original bare-state energies $E_n$. There was no consideration given to dynamically altering the dark state structure — changing the mixing angle — as is required for STIRAP.

It is unfortunate that the comprehensive LASL work [Ohl67] was never published. It is available now on the internet. The LASL work on "spin filters", making use of population trapping, is a derivative of the Lamb work, and it is the latter, particularly [Lam52], that therefore deserves recognition for early discovery of population trapping.

The literature on population trapping [Lam52, Sho77, Gra78, Rad82, Dal82a, Dal82b] and dark states [Ari76, Alz76, Ari76, Alz79] prior to the introduction of STIRAP dealt primarily with

steady illumination, not with pulsed adiabatic following, and so it dealt with dark states that had static components and a fixed mixing angle. Only with the allowance of (adiabatically) changing mixing angle does the dark state become associated with adiabatic population transfer and STIRAP.

The authors of [Sho77] were concerned with adjusting fields to optimize effects on atoms and so they gave no consideration to the steady-state complex-valued susceptibility that accompanies an induced dipole moment and alters the fields, forming the basis of EIT, cf. **Sec.** 5, nor did they consider pulsed changes of the trapping state, the basis of STIRAP.

Earlier literature treating adiabatic passage in two- and three-state systems considered only swept frequencies as contributors to adiabatic change. Work on three-state adiabatic following, such as that of Hioe [Hio83a] and Oreg, Hioe and Eberly [Ore84], cf. **Sec.** 2.2.8, dealt with coherence vectors (generalizations of the Bloch vector) as the constructs that underwent "following". It was these rather than adiabatic states that were proposed as a means of understanding quantum-state changes. That work did consider non-coincident pulses (an essential for STIRAP) but was directed toward consideration of frequency sweeps, a natural extension of the chirped adiabatic passage of two-state systems that traces back to 1932 and LZ-LZS-LZSM, cf. **Sec.** 2.1.3.

The possibility of population transfer by means of anti-intuitive (or counter-intuitive) excitation was first recognized by Oreg, Hioe and Eberly in 1984 [Ore84]. However, the implementation they proposed used swept detuning of simultaneous pulses, an extension to three states of the chirped adiabatic-passage commonly used at that time for two-state transfer. By contrast, STIRAP relies on sequenced amplitudes and constant carrier frequencies. Nevertheless they pointed to the possibility of delayed pulses being advantageous for adiabatic inversion, and they recognized situations (LZSM curve crossings) where "anti-intuitive" time evolution was advantageous. Both of these concepts are essential ingredients of STIRAP.

The papers [Kuk89, Gau90] were the first to combine all the theoretical elements that are essential for STIRAP, as defined in **Sec.** 1.3, and to provide a theory supporting the earlier first measurements of population transfer by stimulated Raman adiabatic passage [Gau88]. They built theory around the adiabatic change of eigenvectors and the following of that motion — notably the dark state — by the statevector. As can be seen from **Sec.** 1.3.5 viewing STIRAP as an example of a torque equation provides an even simpler view of the adiabatic following.

Subsequent work has, in many cases, treated systems differing substantially from those that deal with coherent quantum-state excitation. The dependent variables may not be three quantum-state amplitudes, the independent variable may not be time, and the interactions may not originate with traveling electromagnetic fields. Nonetheless, one may find in the equations – variants of the basic eqns. (1.10) and (1.11) and their solutions — analogs of the STIRAP phenomena, as summarized in the bulleted list above. The consequence of meeting the conditions for STIRAP is robust change of the variables $C_n(t)$ from given initial values at $t = 0$ to prescribed values at $t = T$.

## 8 Acknowledgments

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## A Acronyms

The following set of acronyms and phrases are amongst the many that occur in the literature connected with STIRAP. As will be seen some phenomena has had multiple acronyms, and some acronyms have had multiple meanings. The listed citations are to early or popular uses. Many will be found in Wikipedia or Google ® .

- AAP = Assisted Adiabatic Passage. A second field (see CDF) is imposed to improve adiabaticity. [Dem05, Zha13]. Also vacuum-assisted adiabatic passage (VAAP) [Sie09] and dissipation-assisted adiabatic passage (DAAP) [Mar03].
- AAPT = Assisted Adiabatic Population Transfer. [Kob98, Kur01a].
- AF = Adiabatic Following: continual alignment between the either the Bloch vector with the angular-velocity vector or between the statevector and an adiabatic state (or a fixed superposition of such states). [Gri70, Gri73, All75, Loy77, Sho11]. See **Secs.** 1.2.3 1.3.6 2.2.3.
- APLIP = Adiabatic Passage by Light Induced Potentials. [Vit01a, Vit01b, Sha06, Tsc07].
- AP = Adiabatic Passage: Complete population transfer adiabatically[20]. [Mes62, Vit01a, Vit01b, Sha06, Kra07, Sho11]. See **Sec.** 2.2.5.
- ARP = (optical) Adiabatic Rapid Passage (= RAP): Population transfer produced by adiabatic evolution in a time shorter than an incoherence [Loy74, Gri75]. See **Sec.** 2.2.5.
- AT = Autler-Townes splitting (or effect): A strong $S$ field linked to state 2 induces ac Stark shifts that split the 1-2 resonance obseved by a weak $P$ probe. [Aut55, Zol79]. See **Sec.** 2.4 and 2.4.2.
- AWG = Arbitrary Waveform Generator.
- BAP = Bichromatic Adiabatic Passage: Use of two frequencies to produce AP [Yat03, Amn04, Ele12].
- CAPTA = Chirped Adiabatic Passage by Two-photon Absorption [Che97, Tsc07].
- CDF = Counter-Diabatic Field: This is a second field that, when combined with the first field, generates the desired adiabatic population transfer. It works to undo the effects of nonadiabaticity on the population transfer. [Dem03, Dem05, Rus12, Zha13].
- CHIRAP = CHIRped Adiabatic Passage (= RCAP.) [Bro92, Mel92]. See **Sec.** 4.
- CPT = Coherent (or Complete) Population Transfer [Vit03a, Vit03b, Kyo06, Vas06, Kra07, Ran07].
- CPT = Coherent Population Trapping, when population in a coherent superposition of two stable quantum states is not affected by specific radiation. [Aga93, Mar98, Hal01, Win01, Fle05, Lam06]. See **Sec.** 2.6.2.

[20] An alternative view: any completed adiabatic following.

- CPT = Cyclic Population Transfer: repeated population transfer [Kra01].
- CQED = Cavity Quantum Electrodynamics: individual atoms passing through a single-mode cavity exchange excitation energy with single photons [Har93, Wal06].
- CTAP = Coherent Tunneling by Adiabatic Passage [Gre04, Lon07a].
- DFG = Difference Frequency Generation [Suc08, Suc11, Por12].
- DFG = Deutsche Forschungsgemeinschaft (German Research Foundation).
- DFS = Decoherence-Free Subspace, a portion of a larger Hilbert space that is unaffected by coherence-destroying processes [Zan97, Lid98, Dua98].
- DOR = Double Optical Resonance (= OODR) [Zol79]. See **Sec.** 1.1 2.8.3.
- EIT = Electromagnetically Induced Transparency [Har90,Har97,Mar98,Fle99,Fle05]. See **Sec.** 5.
- DSP = Dark State Polariton: a coherent superposition of field and matter states [Fle02]. See **Sec.** 5.4.
- FVH = Feynman, Vernon and Hellwarth vector-model of two-state coherent excitation, depicting the motion of the Bloch vector as governed by a torque equation [Fey57]. See **Sec.** 2.2.2.
- GSD = Ground-State-Depletion: removal of population from the ground state [Aga06, Rub13].
- LASL - Los Alamos Scientific Laboratory. Later renamed by congressional mandate Los Alamos National Laboratory. See **Sec.** 2.3.7.
- LICS = Laser-Induced Continuum Structure: structure embedded in a continuum by a laser field [Kni84, Kni90]. See **Sec.** 2.5.5.
- LWI = Lasing Without Inversion: a strong control field creates a superposition that leaves gain for state 2 and transparency for state 1. [Ima89, Scu89, Koc92, Har97]. See **Sec.** 5.
- LZ or LZS or LZSM = Landau-Zener-Stückelberg-Majorana theory of two-state adiabatic evolution through diabatic curve crossings. [Lan32a, Lan32b, Zen32, Stu32, Maj32]. See **Sec.** 2.1.3.
- MS = Morris-Shore transformation, converts certain $N$-state linkage patterns to a set of independent two-state systems. [Mor83, Vit03, Kyo06, Ran06b, Sho13]. See **App.** E.3.
- NMOR = Nonlinear Magneto-Optical Rotation : light-intensity-dependent rotation of the polarization plane of linearly polarized light during its propagation through a medium placed in an external magnetic field [Bud02, Pus11].
- NV = Nitrogen-Vacancy center in diamond. These are point defects, each of which consists of a substitutional nitrogen adjacent to a vacancy. [Jel06, Neu08, Zha13, Alv11].

- ODE - Ordinary Differential Equation. See eqn. (1.10).
- ODR = Optical Double Resonance. See **Sec.** 2.8.3.
- OODR = Optical-Optical Double Resonance [Dix60, Got78, Bed78, Ham86, Bra87] . See **Sec.** 2.8.3.
- QIP = Quantum Information Processing: manipulation of quantum states that serve as information elements [Zol05].
- QST = Quantum State Transfer [Bis04, Che13, Hou13].
- QST = Quantum State Tomography [The02, Lvo09, Roz12].
- RAP = Rapid Adiabatic Passage (=ARP). Complete population transfer using coherent adiabatic evolution that is more rapid than the spontaneous emission lifetime [McC57, Sho08, Sho11]. See **Sec.** 2.2.5.
- RCAP = Raman Chirped Adiabatic Passage (= CHIRAP). See **Sec.** 4.
- RESOLFT = REversible Saturable OpticaL Fluorescence Transition [Hel94,Kla01,Rub13].
- RIBAP = Retroreflection-Induced Bichromatic Adiabatic Passage [Yat03].
- RRS = Resonance Raman Spectroscopy: the $P$ field is near resonance with an optically allowed (electric-dipole) transition. See **Sec.** 1.1 and [Wikipedia].
- SAP = Spatial Adiabatic Passage protocol [Neg12, Ben12]. See **Sec.** 6.5.
- SAP = Super-Adiabatic Passage: treats adiabatic following in a two-state system subject to excitation by amplitude-modulated field by means of super-adiabatic basis [Vit03a].
- SARP= Stark-induced Adiabatic Raman Passage [Muk11, Muk14].
- SCRAP = Stark Chirped Rapid Adiabatic Passage [Yat99, Ric00, Yat02a, Yat02b, Yat05, Moh05].
- SEP = Stimulated Emission Pumping: The Stokes field of a Raman process is externally generated, [Kit81, Ham86, He90, Dai94, Ber98, Vit01a, Sho03].
- SFG = Sum Frequency Generation [Lon07b, Wal09, New11, Suc11, Ran12c].
- SLAP = Subwavelength Localization via Adiabatic Passage technique [Mom09, Vis12, Men13].
- SRS = Stimulated Raman Scattering; see **Sec.** 1.1 [She76, Ray79, Ray81, Ray85]. See **Sec.** 1.1.
- STED = STimulated-Emission-Depletion. A STED fluorescence microscope overcomes the diffraction resolution limit by depleting the excited state of the fluorescing molecules in the outer region of the focus [Hel94, Rub13].

- STWM = Simultaneous Three-Wave Mixing processes [Por12, Wei13].
- TDSE = Time-Dependent Schrödinger Equation. See eqns. (1.9) and (1.10).
- Tripod = a four-state fan linkage with three states linked only to a common state, by three fields [Una98a, Una99, The99]. See **Sec.** 6.4.

**STIRAP variants**

- STIRAP = STImulated Raman Adiabatic Passage [Gau90, Ber98].
- STIHRAP = STImulated Hyper-Raman Adiabatic Passage : one or two of the STIRAP interactions $P$ and $S$ are two-photon or multiphoton transitions. [Yat98b, Gue98a, Gue98b, Boh01].
- B-STIRAP = Bright-state (or Backward) Stimulated Raman Adiabatic Passage [Kle07, Kle08, Bei08, Bei11, Cha12].
- D-STIRAP= Detuning-induced Stimulated Raman Adiabatic Passage. The application of STIRAP to the Bloch eqations (as in [Vit06]) [Den12].
- Double-STIRAP : a first transfer ($S$ before $P$) is followed by a return transfer ($P$ before $S$) [Tro03, Osp08, Sho10].
- Double-STIRAP : a first transfer is followed by a second transfer to a fourth state (= sequential STIRAP) [Dan08].
- Triple-STIRAP: A sequence of pulselets moves population back and forth between states 1 and 3, finishing with population in state 3 [Sho10].
- F-STIRAP = Fractional Stimulated Raman Adiabatic Passage [Vit99, Wan04, Vit10, Sho11, Zhe11].
- Sequential-STIRAP: a first STIRAP is followed by a second one to a new final state [Kur01].
- S-STIRAP = Straddle-STIRAP; see **Sec.** 6.4 [Mal97, Vit98b].
- V-STIRAP = Vacuum Stimulated Raman Adiabatic Passage: the $S$ field is that of a cavity vacuum [Kuh10, Vas10, Sho11].

## B Terminology

As in all subjects of continued intellectual endeavor, the study of adiabatic processes in coherent excitation has engendered a great variety of notation and terminology, with differing meanings for common words. This section discusses some of those.

### B.1 States

Regarding terminology: I have here followed the common practice of regarding (Hilbert-space) "vector" and (quantum) "state" as synonyms. Thus "eigenvector" and "eigenstate" are interchangeable terms.

In my writings I have followed the lead of the pioneers of quantum theory and referred to "quantum state" (or simply "state") as the most elementary unit of the theory: information that uniquely defines the most detailed description of a system. For a simple system such as hydrogen, that means a quantum state is defined by a complete list of 5 quantum numbers, say $n, \ell, J, M_J, M_I$. There may occur several quantum states (i.e. several sets of quantum numbers) that have the same energy — the states are degenerate but distinct. The presence of a magnetic field typically removes ("lifts") the degeneracy, so that every quantum state then has a unique energy, and every energy is associated with a unique quantum state.

In writings that refer to quantum structures one finds two viewpoints:

**View 1.** The splitting of a degenerate energy level by a magnetic field produces what I, and many other authors, have called "Zeeman sublevels", or simply "sublevels". That is, in the absence of an externally applied magnetic (or electric) field a given energy ("energy level" or "level") may be associated with a set of degenerate quantum states — degenerate sublevels.

**View 2.** The LASL authors [Ohl67], and many others, write of "substates" (or "magnetic substates" or "Zeeman substates"). This nomenclature implies the existence of a single "state" that is to be further categorized with additional quantum numbers (akin to hidden variables). Those that write of "substates" therefore use this as a synonym for what those of View 1 would call "quantum states" or "Zeeman sublevels".

### B.2 Eigenvalues

The connection $E = \hbar\omega$ between energy $E$ and (angular) frequency $\omega$ introduces $\hbar = h/2\pi$, the rationalized Planck constant or Dirac constant. For typographic simplification many authors treat energy and frequency interchangeably, by assuming units such that $\hbar = 1$.

The time-varying adiabatic eigenvalues $\varepsilon_\mu(t)$ of the RWA Hamiltonian need not have units of energy, and must not be confused with the bare energies $E_n$ associated with the system prior to the introduction of the RWA.

## C Phases

The choice of phases $\zeta_n(t)$ defines a *picture*, and with that the diagonal elements $W_{nn} = \Delta_n$ of the matrix $\mathsf{W}(t)$. The *Schrödinger picture* sets the phases to zero, so the diagonal elements are

the bare energies,

$$\zeta_n(t) = 0 \qquad \Delta_n = E_n/\hbar, \qquad \text{Schrödinger picture.} \tag{C.1}$$

The *Dirac* or *interaction picture* makes the diagonal elements zero by choosing the phases

$$\zeta_n(t) = E_n t/\hbar, \qquad \Delta_n = 0, \qquad \text{Dirac picture.} \tag{C.2}$$

This nullification of all diagonal elements facilitates time-dependent perturbation theory, but leaves an exponential time variation multiplying the Rabi frequencies, cf. eqn. (2.18). The *rotating-wave picture* for the three-state chain takes

$$\zeta_2(t) = \zeta_1(t) + \omega_P t, \qquad \zeta_3(t) = \zeta_2(t) \pm \omega_S t, \qquad \text{rotating-wave picture,} \tag{C.3}$$

with the plus sign for a ladder and the minus sign for a lambda linkage. There remains to be fixed $\zeta_1(t)$, for which two choices are common. For lossless excitation these choices, and the consequent diagonal elements, are

$$\zeta_1(t) \quad = E_1 t/\hbar: \qquad \Delta_1 = 0, \qquad \Delta_2 = \Delta_P, \quad \Delta_3 = \Delta_S \pm \Delta_S, \tag{C.4a}$$

$$\zeta_1(t) \quad = E_2 t/\hbar - \omega_P t: \quad \Delta_1 = -\Delta_P, \quad \Delta_2 = 0, \qquad \Delta_3 = \pm\Delta_S, \tag{C.4b}$$

where the plus sign goes with the ladder linkage and the minus sign goes with the lambda linkage.

With any picture the inclusion of loss obtains from the replacement $\Delta_n \to \Delta_n - \mathrm{i}\Gamma_n/2$.

## D Rate equations; Multiple-photon vs. multiphoton

Incoherent radiative excitation described by rate equations for probabilities,

$$\frac{d}{dt}P_n(t) = \sum_n R_{nm}P_m(t), \tag{D.5}$$

and with constant radiative-transition rates $R_{nm}$ parameterized for two states by Einstein $A$ and $B$ coefficients[21] and (resonant) radiation density $u$,

$$\text{excitation } (1 \to 2)\text{: } R_{21} = B_{12}u, \qquad \text{de-excitation } (2 \to 1)\text{: } R_{12} = B_{21}u + A_{21}, \tag{D.6}$$

envisions photons as acting sequentially to produce a succession of *multiple-photon* absorptions (Einstein coefficient $B_{12}$) or stimulated emissions (coefficient $B_{21}$) and spontaneous emissions (coefficient $A_{21}$). Long-established perturbation theory, often presented with Feynman diagrams, uses the same picture: a succession of time-ordered events, e.g. [Guc74, Cou77]. With laser light sources it becomes possible to observe processes described by *multi-photon* (or *poly-photon*) rate coefficients $R_{nm}$ that are proportional to a higher power than first of radiation intensity. In such situations all of the field photons are regarded as being present simultaneously.

When applied to coherent excitation, and the TDSE, the terminology distinction is that in a *multiple photon* process it is possible to extract populations from intermediate states, whereas in a *multi-photon* process any intermediate states are far from resonance and hence are not observable as final states.

[21] The subscript ordering on $A$ and $B$ is conventional and opposite to that used for the matrix $R_{nm}$. The Einstein coefficients are defined for use with energy density $u$ rather than with intensity $I$. When used with $I$, as is appropriate for radiation beams, they are known as Milne coefficients.

# E Dark states and population trapping

## E.1 The dark-state eigenvalue

The existence of an adiabatic state that lacks state-2 component, for the three-state chain with two-photon resonance, is readily seen from matrix multiplication:

$$\begin{bmatrix} a & P^* & 0 \\ P & b & S \\ 0 & S^* & a \end{bmatrix} \begin{bmatrix} S \\ 0 \\ -P \end{bmatrix} = a \begin{bmatrix} S \\ 0 \\ -P \end{bmatrix}. \tag{E.7}$$

In general all of the elements $a, b, P, S$ may be time dependent. As can be seen, the 1,1 and 3,3 elements of the matrix must be equal in order for the dark state to be an eigenvector. This is the condition for two-photon resonance. The common association of the dark state with the null-value eigenstate rests on a choice of phases $\zeta_n$ that nullifies the diagonal element $a$, cf. eqn. (C.4a).

## E.2 A simple example of population trapping

A simple example of population trapping can be seen in a model of excitation by linearly polarized light acting on a degenerate two-level system [Sho76, Sho90, Sho11]. When the quantization axis, $(z)$, is taken along the polarization direction then the linkage pattern is as shown in **Fig.** E.1$(a)$. The population initially present in Zeeman sublevel $J = 1, M = 0$ will Rabi cycle into the excited state, $J = 0, M = 0$, from which it will be lost, say by photoionization. Eventually only the two sublevels $M = \pm 1$ will remain populated: 1/3 of the initial population will be removed.

If, however, we choose the quantization axis to be along the beam propagation direction, and hence perpendicular to the polarization direction, as show in the inset of frame $(b)$, then the field acting on the system must be expressed as a linear superposition of right- and left-circular polarization. The selection rules for dipole radiation are now $\Delta M = \pm 1$ rather than the $\Delta M = 0$ of frame $(a)$, and the linkage pattern is that of frame $(b)$. It would appear that this relabeling of coordinates will remove 2/3 of the initial population. This intuition is wrong: the system will be left in a coherent superposition of the $M = \pm 1$ sublevels — a dark state.

In this example the trapped state occurs because a rotation of coordinates alters the sublevel composition of an angular momentum state: what is seen as a single magnetic sublevel in one reference frame will appear as a coherent superposition in another frame [Zar88, Sho90]. Intuition based on rate-equations, as is appropriate for incoherent excitation, cannot deal with coherent superpositions.

## E.3 Multiple dark states

For many $N$-state linkage patterns, such as the resonant fan (e.g tripod) or chain (e.g. letter-M) there exist multiple dark states — superpositions of the basic bare states that are unaffected by the interaction. The analysis of such situations proceeds by partitioning the $N$ states into two classes, $N_A$ in set $A$ and $N_B$ in set $B$, such that there are no interactions within a class, only

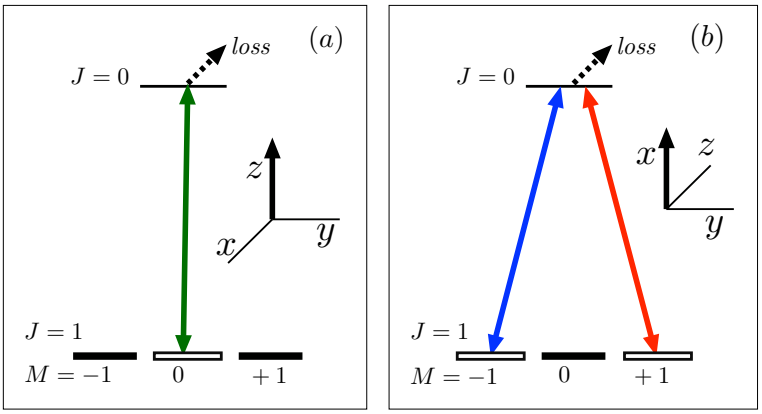


Fig. E.1. $(a)$ Excitation of degenerate ground level, $J = 1$ to a lossy excited state $J = 0$ by radiation linearly polarized along quantization axis $z$. $(b)$ Linkage pattern for linear polarization expressed as coherent superposition of right- and left-circular polarization. Insets show geometry of polarization vector (bold arrow). The boxes for the $J = 1$ sublevels indicate states from which population is removed. In frame $(a)$ the removal is complete. In frame $(b)$ only half the population is removed.

between state of different classes. The sets $A$ and $B$ may differ in size but all states are members of one of these sets. Let the smaller number be denoted by $N_<$:

$$N = N_A + N_B, \qquad N_< = \min(N_A, N_B). \tag{E.8}$$

I shall assume that the set $A$ contains the initial state and the set $B$ contains at least one of the excited states. Let the system must be such that, with this partitioning and a suitable ordering of the quantum states (all $A$ states preceding all $B$ states in listings), the instantaneous RWA Hamiltonian matrix has the structure

$$\mathsf{W} = \begin{bmatrix} \Delta_A \mathsf{1}_A & \mathsf{V} \\ \mathsf{V}^\dagger & \Delta_B \mathsf{1}_B \end{bmatrix}, \tag{E.9}$$

where $\Delta_A$ and $\Delta_B$ are scalars, $\mathsf{1}_A$ and $\mathsf{1}_B$ are square unit matrices, of dimension $N_A$ and $N_B$ respectively, $\mathsf{V}$ is a rectangular matrix, of dimension $N_A \times N_B$, and $\mathsf{V}^\dagger$ is its Hermitian conjugate (complex conjugate of transpose). That is, in the RWA the $A$ states are degenerate, as are the $B$ states, and there are no transitions within these sets. Note that it is always possible, by suitable choice of the phases $\zeta_n(t)$ used in defining the probability amplitudes $C_n(t)$, to nullify the $A$ detuning, $\Delta_A = 0$. With this choice then when the transitions are all resonant both detunings will vanish, $\Delta_A = \Delta_B = 0$, a commonly treated situation.

For such a linkage pattern it is possible to introduce a new set of Hilbert-space coordinates, using a Morris-Shore transformation [Mor83, Sho13]

$$\widetilde{C}_j(t) = \sum_j M_{jn} C_n(t) \tag{E.10}$$

that produces the TDSE

$$\mathrm{i}\frac{d}{dt}\widetilde{\mathbf{C}}(t) = \widetilde{\mathsf{W}}(t)\widetilde{\mathbf{C}}(t), \tag{E.11}$$

in which the transformed RWA Hamiltonian matrix $\widetilde{\mathsf{W}} = \mathsf{MWM}^\dagger$ is block diagonal, i.e. along the diagonal are a set of $2 \times 2$ blocks $\mathsf{w}^{(n)}$ together with a unit matrix $1_U$ of dimension $N_U$,

$$\widetilde{\mathsf{W}} = \begin{bmatrix} \mathsf{w}^{(1)} & 0 & 0 & \cdots & 0 \\ 0 & \mathsf{w}^{(2)} & 0 & \cdots & 0 \\ 0 & 0 & \mathsf{w}^{(3)} & \ddots & 0 \\ \vdots & \vdots & \vdots & \ddots & 0 \\ 0 & 0 & 0 & 0 & \Delta 1_U \end{bmatrix}. \tag{E.12}$$

The matrices $\mathsf{w}^{(n)}$ (there are $N_<$ of these) have the structure

$$\mathsf{w}^{(n)} = \begin{bmatrix} \Delta_A & \frac{1}{2}\widetilde{\Omega}^{(n)} \\ \frac{1}{2}\widetilde{\Omega}^{(n)} & \Delta_B \end{bmatrix}, \tag{E.13}$$

in which every block has the same pair of detunings as the original system, $\Delta_A$ and $\Delta_B$, but the Rabi frequencies $\widetilde{\Omega}^{(n)}$ may differ from block to block. The $\Delta$ associated with the unit matrix $1_U$ is the detuning associated with the larger set of states: it is $\Delta_A$ if $N_A > N_B$ and is $\Delta_B$ if $N_B > N_A$. It is absent if $N_A = N_B = N_<$, in which case all the elements of $\widetilde{\mathsf{W}}$ describe pairs of interacting states.

The MS transformation replaces the $N$ linked states by a set of $N_<$ coupled pairs, one for each member of the smaller set of states. Any remaining states, $N_U$ in number, are unpaired and unaffected by the interactions $\mathsf{V}$ and $\mathsf{V}^\dagger$. This number is

$$N_U = |N_A - N_B| = N - 2N_<. \tag{E.14}$$

If the unlinked components are in the $A$ set, and hence they are unlinked from excited states in set $B$, they are dark states or population-trapping states. If the components of the unpaired state are in the $B$ set they are spectator states.

This simple enumeration shows that the resonant tripod linkage has two dark states [Una98a], the letter-M has one and the letter-W has one spectator state. As with the single dark state of the lambda linkage, the structure of these states changes as the RWA Hamiltonian changes.

## F Autler-Townes splitting

### F.1 AT and EIT

The work of Autler and Townes [Aut55] treated a completely coherent model of a two-state atom subject to excitation by a monochromatic field, with and without what is now called the rotating-wave approximation. They used Floquet theory to find the frequencies with which the probability amplitudes oscillated, a pair of values separated by what is now termed the Rabi frequency. These form the *AT doublet*, separated by the AT splitting.

Their model had no spontaneous emission or other sources of line width. They did not examine details of the wave function that would be revealed by a weak probe linkage to a third state, most notably the destructive interference that gives the notable transparency now associated with electromagnetically induced transparency and discussed by Harris [Har97].

Because the EIT profile, as found when the two strongly coupled states decay into a single continuum, has an interference null, it differs from a superposition of two Lorentzian profiles where no such null occurs. In recent years the term "AT profile" has been used for such profiles [Ani11], to distinguish them from EIT profiles that have a Fano minimum, although the original AT model had no such incoherence-based profiles.

The observation of a null in the probed transition to two strongly coupled states is properly termed EIT. The presence of two peaks in an observed spectral profile, whether or not there occurs transparency, is properly termed an Autler-Townes effect, necessarily present in any EIT.

The Fano minimum of EIT occurs even with a small AT splitting. However, its observation then requires a long time, and so it may not be apparent with pulsed excitation. When the two strongly coupled states each undergo spontaneous emission, there are two continuua, and the null does not occur.

### F.2 Autler-Townes states

To treat the combination of a weak $P$ field and a strong $S$ field, $|\Omega_S| \gg |\Omega_P|$, we introduce dressed states of the strongly coupled 2-3 subsystem. The original equations (1.27) (not restricted to two-photon resonance but with a stable state 3, so $\Gamma_3 = 0$) become

$$\mathrm{i}\frac{d}{dt}\begin{bmatrix} C_1(t) \\ A_-(t) \\ A_+(t) \end{bmatrix} = \tfrac{1}{2}\begin{bmatrix} 0 & \Omega_- & \Omega_+ \\ \Omega_- & 2\Delta_P - \mathrm{i}2\Gamma_2 \pm \Delta_S - \delta & 0 \\ \Omega_+ & 0 & 2\Delta_P - \mathrm{i}2\Gamma_2 \pm \Delta_S + \delta \end{bmatrix}\begin{bmatrix} C_1(t) \\ A_-(t) \\ A_+(t) \end{bmatrix}, \tag{F.15}$$

where the sign $+$ goes with the ladder and the $-$ sign goes with the lambda linkage and $\delta$ is

$$\delta = \sqrt{(\Delta_S)^2 + |\Omega_S/2|^2}. \tag{F.16}$$

Each of the two dressed states acquires the decay rate $\Gamma_2$ of state 2.

**Resonant $P$.** Let the $S$ field be very strong and resonant with the 2-3 transition, $\Delta_S = 0$. Then if the $P$ field maintains its resonant detuning with state 2, $\Delta_P = 0$, there will occur little excitation from state 1 with steady illumination: the population remains trapped there, making no transition into either dressed state. The larger is $|\Omega_S|$ the more complete is the population trapping. When $|\Omega_S|$ is not much larger than $|\Omega_P|$ population regularly leaves state 1 for states 2 and 3.

**Figure** 2.14 of **Sec.** 2.9.2 shows examples of this behavior, for a lossless system with $\Delta_S = 0$. In frame $(a)$ the $S$ field is weaker than the $P$ field, and the dynamics is dominated by Rabi oscillations between states 1 and 2. As the $S$ field grows stronger, and $\Delta_P = 0$, population increasingly remains in state 1 — it becomes trapped there.

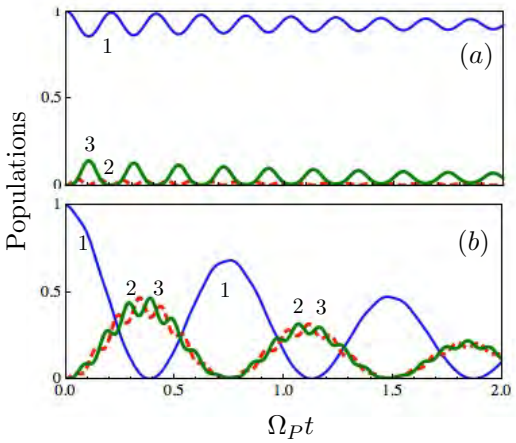


Fig. F.2. Example of three-state excitation with constant intensity and strong resonant $S$ field, $\Delta_S = 0$, $\Omega_S = 5\Omega_P$, for different $P$-field detunings and loss rate $\Gamma_2 = 0.1\Omega_P$. $(a)$ $\Delta_P = 0$: $P$ field resonant with state 2. $(b)$ $\Delta_P = 0.5\Omega_S$: $P$ field resonant with dressed state $|+\rangle$. Populations of states 2 and 3 are intertwined; they are equal components of $|+\rangle$. All states decay equally.

**Detuned $P$.** With nonzero values of $\Delta_P$ the statevector will have a bright-state component, and this will undergo excitation into lossy state 2. When the pulses persist for many lifetimes of state 2 then the bright component will produce $P$-field attenuation and fluorescence.

State 1 will be resonant with one of the two AT dressed states (and thereby undergo maximum excitation and loss) when the real part of a second diagonal element of $\mathsf{W}(t)$ is zero. When $\Delta_S = 0$ that will occur with the $P$-field detuning $\Delta_P = \pm|\Omega_S/2|$. This is half the Autler-Townes splitting. Frame $(c)$ of **Fig.** 2.14, making this choice, shows the resulting Rabi oscillations between state 1 and the AT dressed state $|+\rangle$.

**Figure** F.2 shows two examples of population histories when there is loss from state 2 and a strong resonant $S$ field ($\Omega_S = 5\Omega_P$ and $\Delta_S = 0$), for two values of the $P$-field detuning $\Delta_P$. When the $P$ field and the $S$ field are both resonant with their bare-state transitions, frame $(a)$, the loss has little effect on the population, which remains basically trapped in state 1. As the $S$ field becomes stronger the trapping becomes more complete. When the $P$ field is tuned to a dressed-state resonance, as in frame $(b)$, there occur damped Rabi oscillations into a superposition of states 2 and 3.

## G The adiabatic condition

The condition for adiabatic evolution (and AF) can be found by expressing the original probability amplitudes in an adiabatic basis,

$$\mathbf{C}(t) = \sum_{\mu=+,0,-} A_\mu(t)\mathbf{\Phi}_\mu(t). \tag{G.17}$$

The resulting equations, with assumed two-photon resonance $\Delta_3 = 0$, read (with suppression of explicit time dependence) [Vit01a]

$$\mathrm{i}\frac{d}{dt}\begin{bmatrix} A_+ \\ A_0 \\ A_- \end{bmatrix} = \begin{bmatrix} \frac{1}{2}\Omega_{\rm rms}\,\cot\varphi & \mathrm{i}\dot\theta\,\sin\varphi & \mathrm{i}\dot\varphi \\ -\mathrm{i}\dot\theta\,\sin\varphi & 0 & -\mathrm{i}\dot\theta\,\cos\varphi \\ -\mathrm{i}\dot\varphi & \mathrm{i}\dot\theta\,\cos\varphi & \frac{1}{2}\Omega_{\rm rms}\,\tan\varphi \end{bmatrix}\begin{bmatrix} A_+ \\ A_0 \\ A_- \end{bmatrix}, \tag{G.18}$$

where

$$\tan\theta = \frac{\Omega_P}{\Omega_S}, \qquad \tan 2\varphi = \frac{\Omega_{\rm rms}}{\Delta}. \tag{G.19}$$

The condition for adiabatic following is that the matrix of coefficients be close to diagonal, meaning that the non-adiabatic off-diagonal elements, here involving time derivatives of $\theta(t)$ and $\varphi(t)$, should be much smaller than the separation of the diagonal elements, i.e. the adiabatic eigenvalues. In the limit of vanishing off-diagonal elements there occurs "perfect adiabatic evolution" (an idealization akin to the frictionless plane), with $A_\mu(t)$ changing only by a phase. The non-adiabatic coupling term $\dot\varphi$ vanishes when $\Delta = 0$ (the eigenvalue separation is then $\Omega_{\rm rms}$) but it couples $A_+$ and $A_-$ when there is nonzero single-photon detuning. In the limit of large single-photon detuning this adiabatic basis is inappropriate; see **Sec.** 2.10.1.

When pulses rather than steady fields are used, the appropriate approximation to the probability amplitudes used for adiabatic following are [Vit01a]

$$C_1 = \cos\theta = \frac{\Omega_S}{\Omega_{\rm rms}}, \qquad C_3 = -\sin\theta = -\frac{\Omega_P}{\Omega_{\rm rms}}, \qquad C_2 = \frac{2\mathrm{i}}{\Omega_{\rm rms}}\,\dot\theta. \tag{G.20}$$

Only in the limit of very slow change in mixing angle $\theta$ is there no population in the decaying state.

The condition for adiabatic evolution in STIRAP has been expressed in three ways, which I shall call : "local", "worst-case" and "global".

**Local condition.** The first of these comes from the requirement that change of the RWA Hamiltonian $\mathsf{W}(t)$ be very slow. For the STIRAP system this becomes a requirement that the change in mixing angle $\theta(t)$ be much less than the eigenvalue separation, which for fully resonant excitation ($\Delta = 0$) is the rms Rabi frequency:

$$\dot\theta(t) \ll \Omega_{\rm rms}(t), \tag{G.21}$$

For resonant excitation this translates into condition (G.22) on changes of the Rabi frequencies. This is a "local condition" for adiabatic evolution, and it must be satisfied at all times. The pulses must therefore be smooth, with no rapid variations. For fully resonant excitation the adiabatic condition can be written

$$\left|\Omega_P\dot\Omega_S - \Omega_S\dot\Omega_P\right| \ll \left|\Omega_{\rm rms}\right|^3. \tag{G.22}$$

We recognize that when either field is very small, there can be abrupt changes in the other field without violating this condition.

**Worst-case condition.** The early papers on STIRAP [Kuk89, Gau90] dealt with Gaussian pulses of equal peak Rabi frequency and Gaussian width $T$. They examined the requirement that at the midpoint of the pulse sequence, $t = 0$, where the adiabatic condition was most restrictive, the evolution would satisfy the local condition (G.21), and they found, for optimally delayed pulses, an inequality that can be written

$$\Omega_{\rm rms}(0)\, T \gg 1. \tag{G.23}$$

This is a "worst-case condition" of eqn. (G.21). It was expected to be applicable not only for Gaussian pulses but for other optimally delayed smooth shapes.

**Global condition.** A "global condition" on the Rabi frequencies is obtainable by integrating eqn. (G.21). The integral of the mixing angle from its initial value of 0 (dominant $S$ field) to its final value of $\pi/2$ (dominant $P$ field) is, for any form of intermediate time variation $\pi/2$, The integral of a Rabi frequency $\Omega_n(t)$ is a (temporal) pulse area $\mathcal{A}_n$. Thus by integrating eqn. (G.21) we obtain the global condition that the rms pulse area be much larger than $\pi/2$,

$$\mathcal{A}_{\rm rms} \gg \pi/2. \tag{G.24}$$

This inequality has often been replaced by the statement that the individual pulse areas must be larger than 10 (or $3\pi$) [Ber98, Vit01b], but see **Sec.** 1.3.7.

## H Simulation

In my simulations I express the Rabi frequencies in terms of pulse envelopes $f(t)$ whose peak values are unity:

$$\Omega_P(t) = \Omega_{P0}\, f_P(t), \qquad \Omega_S(t) = \Omega_{S0}\, f_S(t). \tag{H.25}$$

The two envelopes are characterized by widths $\tau_P, \tau_S$ (usually taken to be equal, $\tau_P = \tau_S = \tau$ ) and center-moments $t_P, t_S$, offset in time by delay $t_P - t_S$,

$$f_P(t) = f\left(\frac{t - t_P}{\tau_P}\right), \qquad f_S(t) = f\left(\frac{t - t_S}{\tau_S}\right). \tag{H.26}$$

Usually the pulses have the same peak values, $\Omega_P = \Omega_S = \Omega$. The present simulations used two shapes for $f(t)$, a Gaussian $f_e(x)$ and a finite-duration cosine-squared $f_c(x)$,

$$f_e(x) = \exp(-x^2), \qquad f_c(x) = \begin{cases} \cos^2(x), & |x| < \pi/2, \\ 0 & |x| \geq \pi/2. \end{cases} \tag{H.27}$$

I solve the resulting coupled equations (1.27) using the `NDSolve` macro of Mathematica ® . For steady fields the time interval runs from 0 to $T$, while for pulses it runs from $-T/2$ to $+T/2$, with $t = 0$ at the center of the pulse sequence.

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