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WINTER COLLEGE ON LASERS, ATOMIC AND MOLECULAR PHYSICS

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Optical Coherent Transients and Pulse Propagation

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1. Introduction

These lectures review a new branch of optical spectroscopy, what may be called *coherent optical spectroscopy.* Due to the availability of coherent laser light, atomic and molecular quantum states can now be prepared in coherent superposition, in the same way that nuclear spin systems have been prepared for over 25 years in pulsed nuclear magnetic resonance [1]. Coherently prepared samples of this type exhibit a class of transient phenomena that offer new ways to obtain ultra high resolution optical spectra, and they allow the optical spectroscopist to isolate for the first time individual relaxation processes that have remained hidden within the optical lineshape. Many of these optical transients have now been realized using the recently introduced Stark switching technique [2] and will be reviewed.

We shall treat these coherent transient effects semiclassically using the coupled Schrödinger-Maxwell equations. Our procedure is to first solve the Schrödinger equation, either in density matrix or Bloch form, for a molecular system subject to a pulsed or continuous wave coherent optical field. This preparative stage generates a sample polarization that can radiate coherent light even after the preparation has passed, as prescribed by Maxwell's equations. Each transient effect corresponds to a particular sequence of one or more preparative steps, followed by periods where the sample freely radiates and where the polarization solutions can be traced from one stage to the next. In the case of a gas, the final polarization must be averaged over the Doppler velocity distribution. As long as the radiated field amplitude is small compared to the applied laser field, as in the present measurements, the solution is self-consistent and the laser field can be assumed constant; this is the so-called thin sample regime. At high optical densities, other coherence phenomena such as self-induced transparency [3] occur.

1.1. Density matrix equations

We consider a molecular two-level quantum configuration where the lower level is labeled 1 and the upper level is 2. In sect. 3, the corresponding three-level problem is treated. The molecular gas sample encounters a laser field

$$E_x(z,t) = E_0 \cos(\Omega t - kz) \tag{1.1}$$

of frequency Ω and amplitude E_0, polarized along the x axis and propagating in the z direction. The time-dependent behavior of the density matrix ρ is given by the Schrödinger equation of motion

$$i\hbar\dot{\rho} = [H, \rho] + \text{relaxation terms}\,. \tag{1.2}$$

Lecture Notes from Course at Princeton University, 1977–1978, by H. Gibbs

OPTICAL COHERENT TRANSIENTS & PULSE PROPAGATION

References: chapters in Yariv; Sargent, Scully, + Lamb; Allen + Eberly
R.G. Brewer, "Coherent Optical Spectroscopy," Les Houches, Session XXVII 6/30–7/26, 1975 in R. Balian, S. Haroche, S. Liberman, Frontiers in Laser Spectroscopy, Vol. 1 (North-Holland, Amsterdam, 1977).

The Hamiltonian

$$H = H_0 + H_I \tag{1.3}$$

contains the free molecule part H_0 where the eigenenergies are

$$\langle 1|H_0|1\rangle = \hbar\omega_1 \,,$$

$$\langle 2|H_0|2\rangle = \hbar\omega_2 \,, \tag{1.4}$$

and the 1–2 level splitting in angular frequency units is

$$\omega_2 - \omega_1 = \omega_{21} \,. \tag{1.5}$$

The molecule-optical field interaction term

$$H_I = -\mu \cdot E_x(z, t) \tag{1.6}$$

has the electric dipole matrix element

$$\langle 1|\mu_x|2\rangle = \mu_{12} \neq 0 \,. \tag{1.7}$$

For this two-level problem, eq. (1.2) becomes

$$\dot\rho_{11} = i\chi(\rho_{21} - \rho_{12})\cos(\Omega t - kz) - (\rho_{11} - \rho_{11}^0)/T_1 \,, \tag{1.8a}$$

$$\dot\rho_{12} = i\chi(\rho_{22} - \rho_{11})\cos(\Omega t - kz) - (i\omega_{12} + 1/T_2)\,\rho_{12} \,, \tag{1.8b}$$

where we have introduced the phenomenological decay time T_1, assumed equal for the diagonal elements ρ_{11} and ρ_{22}, and T_2 for ρ_{12}. The source term ρ_{11}^0 denotes that molecules enter level 1 via relaxation from other quantum states and we have defined the Rabi flopping frequency as

$$\chi = \mu_{12}E_0/\hbar \,. \tag{1.9}$$

A similar equation in $\dot\rho_{22}$ can be obtained from (1.8a) by the index interchange $1 \leftrightarrow 2$ and an equation in $\dot\rho_{21}$ from (1.8b) by taking its complex conjugate.

The rapidly oscillating factors of the off-diagonal elements can be removed with the substitution

$$\rho_{12} = \tilde\rho_{12}\mathrm{e}^{i(\Omega t - kz)} \,, \tag{1.10}$$

and by neglecting non-resonant high frequency terms that oscillate as $\mathrm{e}^{2i(\Omega t - kz)}$. This procedure, known as the rotating wave approximation, reduces (1.8) to

NA

$$\frac{\mathrm{d}\rho_{11}}{\mathrm{d}t} = \tfrac{1}{2}i\chi(\tilde\rho_{21} - \tilde\rho_{12}) - (\rho_{11} - \rho_{11}^0)/T_1 \,, \tag{1.11a}$$

$$\left(\frac{\mathrm{d}}{\mathrm{d}t} - i\Delta + \frac{1}{T_2}\right)\tilde\rho_{12} = \tfrac{1}{2}i\chi(\rho_{22} - \rho_{11}) \,, \tag{1.11b}$$

where

$$\Delta = -\Omega + kv_z + \omega_{21} \,. \tag{1.12}$$

We also have recognized that the time derivative

$$\dot\rho = \left(\frac{\partial}{\partial t} + v_z\frac{\partial}{\partial z}\right)\rho \tag{1.13}$$

includes a second term because of a molecular velocity component v_z along the z axis that results in the Doppler shift kv_z of (1.12). Note that at resonance $\Delta = 0$.

1.2. Bloch equations

It is sometimes convenient to work with combinations of (1.11) and the corresponding equations for $\dot\rho_{22}$ and $\dot{\tilde\rho}_{21}$. For example, we may obtain

$$\dot{\tilde\rho}_{12} + \dot{\tilde\rho}_{21} = -i\Delta(\tilde\rho_{21} - \tilde\rho_{12}) - (\tilde\rho_{12} + \tilde\rho_{21})/T_2 \,, \tag{1.14a}$$

$$i(\dot{\tilde\rho}_{21} - \dot{\tilde\rho}_{12}) = \Delta(\tilde\rho_{12} + \tilde\rho_{21}) + \chi(\rho_{22} - \rho_{11}) - i(\tilde\rho_{21} - \tilde\rho_{12})/T_2 \tag{1.14b}$$

$$\dot\rho_{22} - \dot\rho_{11} = -i\chi(\tilde\rho_{21} - \tilde\rho_{12}) - (\rho_{22} - \rho_{11})/T_1 + (\rho_{22}^0 - \rho_{11}^0)/T_1 \,. \tag{1.14c}$$

These are the Bloch equations and may be written as

$$\dot u + \Delta v + u/T_2 = 0 \,, \tag{1.15a}$$

$$\dot v - \Delta u - \chi w + v/T_2 = 0 \,, \tag{1.15b}$$

$$\dot w + \chi v + (w - w^0)/T_1 = 0 \,, \tag{1.15c}$$

using the variables

$$u = \tilde\rho_{12} + \tilde\rho_{21} \,, \tag{1.16a}$$

$$v = i(\tilde\rho_{21} - \tilde\rho_{12}) \,, \tag{1.16b}$$

$$w = \rho_{22} - \rho_{11} \,. \tag{1.16c}$$

The set of eqs. (1.15) can be written compactly as

$$\frac{\mathrm{d}B}{\mathrm{d}t} = \boldsymbol\beta \times B \tag{1.17}$$

where these vectors have components

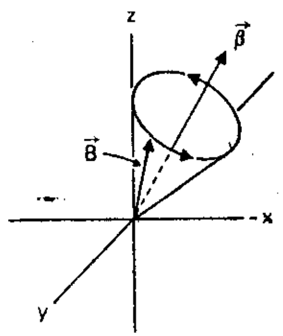
$$B = iu + jv + kw = [u, v, w] \, , \tag{1.18}$$

$$\beta = i\chi + k\Delta = [-\chi, 0, \Delta] \, , \tag{1.19}$$

and we have omitted relaxation for the moment [4]. Eq. (1.17) has the geometric interpretation of a vector of constant length B, the Bloch vector, precessing about an effective field β (fig. 1). In the nuclear magnetic resonance case formulated by Bloch [5], the spins precess in real space about an effective magnetic field, consisting of a static and an oscillating part. In the electric dipole or optical case considered here, the precession is not in real space, but is only a geometric interpretation of a mathematical result. Relaxation may be included simply in (1.17) if we let $T = T_2 = T_1$, and thus the Bloch vector shrinks with time as $e^{-t/T}$. In general, the Bloch vector will not only be a function of z and t but of other variables such as molecular velocity v, namely, $B(v, z, t)$.

The solutions to eq. (1.15), which were discussed initially by Torrey [6] are of the form

$$M(t) = A e^{-at} + B e^{-bt} \cos st + C e^{-bt} \sin st + D \, , \tag{1.20}$$



$$\frac{d\vec{B}}{dt} = \vec{\beta} \times \vec{B}$$

$$\vec{B}(t) = [u(t), v(t), w(t)]$$

$$\vec{\beta} = [-\chi, 0, \Delta]$$

Fig. 1. The Bloch vector B in its precessional motion about the effective field $\vec{\beta}$ provides a simple geometrical representation of the time-dependent behavior of the molecule-optical field interaction, following directly from Schrödinger's equation.

where $M(t) = u(t)$, $v(t)$ or $w(t)$. The D term expresses steady-state behavior and the other terms are transient in nature. The constants cannot be obtained analytically in general except for certain specialized cases such as

$$\Delta = 0 \, ,$$

$$T_1 = T_2 \, ,$$

$$\chi \gg 1/T_1 \, , \qquad \chi \gg 1/T_2 \, .$$

1.3. Polarization and field equations

The light wave, eq. (1.1), induces in a sample of molecular density N the polarization

$$P(z, t) = N\mu_{12} e^{i(\Omega t - kz)} \langle \tilde{\rho}_{12} \rangle + \text{c.c.} \tag{1.21}$$

The bracket $\langle \, \rangle$ denotes an average over the Maxwellian molecular velocity distribution

$$\langle \tilde{\rho}_{12} \rangle = \frac{1}{ku\sqrt{\pi}} \int_{-\infty}^{\infty} \tilde{\rho}_{12} e^{-(\Delta/ku)^2} \, d\Delta \, , \tag{1.22}$$

where Δ is given by (1.12) and u is the rms velocity, not to be confused with the Bloch vector component. Note that $\tilde{\rho}_{12} = \frac{1}{2}(u + iv)$. This polarization, in turn, is the origin of a signal field

$$E_s(z, t) = E_{12}(z, t) \, e^{i(\Omega t - kz)} + \text{c.c.} \, , \tag{1.23}$$

which can be calculated from Maxwell's equations

$$\frac{\partial E_{12}}{\partial z} = -2\pi i k N \mu_{12} \langle \tilde{\rho}_{12} \rangle \, . \tag{1.24}$$

In eq. (1.24), only the lowest order terms of the wave equation

$$\frac{\partial^2 E(z, t)}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E(z, t)}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 P(z, t)}{\partial t^2} \tag{1.25}$$

have been retained, a procedure that is justified as long as the signal amplitude is much smaller than the laser field, $E_{12} \ll E_0$.

1.4. Optical nutation

Simplified case. Consider now the transient effect that results when a molecular sample is suddenly exposed to intense resonant laser light. Molecules begin to execute an alternating absorption and re-emission of laser light as

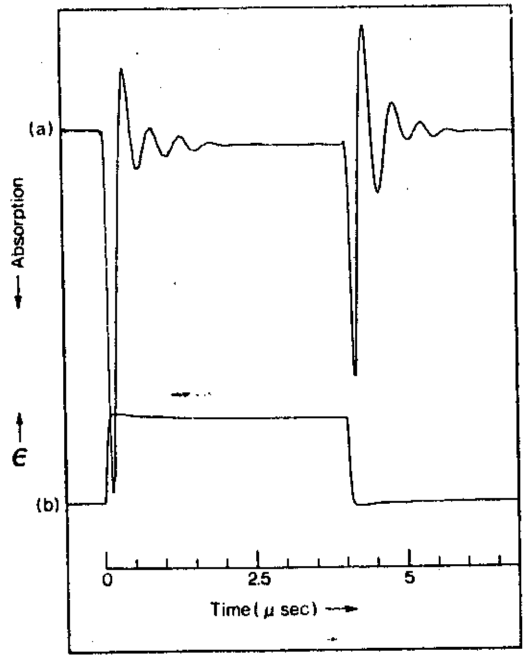


Fig. 2. The optical nutation effect in $^{13}CH_3F$ following a Stark pulse of amplitude ϵ = 35 V/cm. (From Brewer and Shoemaker [2].)

they are driven coherently between upper and lower states (fig. 2). The effect has been called "optical nutation" [7] by analogy with Torrey's transient "spin nutation" [6].

To illustrate the solution of the Bloch equations for this effect, we first simplify the problem by omitting the decay terms in (1.15) so that

$$\dot{u} + \Delta v = 0 \,, \tag{1.26a}$$

$$\dot{v} - \Delta u - \chi w = 0 \,, \tag{1.26b}$$

$$\dot{w} + \chi v = 0 \,. \tag{1.26c}$$

These are easily solved to give in the regime $t > 0$,

$$u(t) = \frac{\Delta w(0)}{\beta^2} (\cos \beta t - 1) \,, \tag{1.27a}$$

$$v(t) = \frac{\chi w(0)}{\beta} \sin \beta t \,, \tag{1.27b}$$

$$w(t) = w(0) \left[1 + \frac{\chi^2}{\beta^2} (\cos \beta t - 1)\right] , \tag{1.27c}$$

where the initial conditions at time $t = 0$,

$$B(0) = [0, 0, w(0)] \,, \tag{1.28}$$

have been utilized. Eq. (1.27) states that the amplitude of the Bloch vector

$$B(t) = [u^2(t) + v^2(t) + w^2(t)]^{1/2} = w(0) \tag{1.29}$$

is preserved at all times, and that it precesses about the effective field β of (1.19) with a frequency

$$\beta = \sqrt{\Delta^2 + \chi^2} \,. \tag{1.30}$$

This motion is especially easy to visualize for the case of exact resonance, when $\Delta = 0$, because $u(t) = 0$ and the Bloch vector executes a right angle rotation about $\beta = \chi$; see fig. 1.

We now apply the results of subsect. 1.3. According to (1.24), the amplitude of the signal field which exits a sample of length L is

$$E_{12}(L, t) = -2\pi i k N L \mu_{12} \langle \tilde{\rho}_{12} \rangle \,, \tag{1.31}$$

and the Doppler-averaged density matrix is given by (1.22),

$$\begin{aligned} \langle \tilde{\rho}_{12} \rangle &= \frac{1}{ku\sqrt{\pi}} \int_{-\infty}^{\infty} \frac{1}{2} (u + iv)\, e^{-(\Delta/ku)^2}\, d\Delta \\ &\approx \frac{i\chi w(0)}{ku\sqrt{\pi}}\, e^{-(\Delta_1/ku)^2} \int_0^{\infty} \left(\frac{\sin \sqrt{\Delta^2 + \chi^2}\, t}{\sqrt{\Delta^2 + \chi^2}} \right) d\Delta \\ &\approx \frac{i\sqrt{\pi}}{2ku}\, \chi w(0)\, e^{-(\Delta_1/ku)^2} J_0(\chi t) \,. \end{aligned} \tag{1.32}$$

For simplicity, we have assumed that the excitation is centered near the Doppler peak at $\Delta = \Delta_1$ so that the Bloch component $\langle u \rangle \sim 0$. Furthermore, we assume that the bandwidth excited is narrow compared to the Doppler width ku so that the Gaussian may be taken outside the integral. Combining (1.31) and (1.32), we obtain for the signal field (1.23)

$$E_s(L, t) = \frac{2\pi^{3/2}}{u} (NL)\, \chi w(0)\, \mu_{12} e^{-(\Delta_1/ku)^2} J_0(\chi t) \cos(\Omega t - kL) \,. \tag{1.33}$$

The transmitted laser beam thus exhibits a slow oscillation, of frequency $\sim\chi$, that is expressed by the zero-order Bessel function $J_0(\chi t)$ and reflects the rate that molecules are driven between lower and upper states. This is essentially the result that is obtained in a more complete treatment where damping is not neglected, the case we will consider next. Fig. 2 shows the "nutation" effect which displays the Bessel function ringing behavior [2].

Nutation with damping. We now give the results of a similar calculation [7,8] where the damping terms in (1.15) are not neglected. Since the Bloch equations did not yield to an analytical solution in general, we need to assume that $T = T_2 = T_1$. We find in a straightforward manner that for times $t > 0$

$$
\begin{aligned}
u(t) = \mathrm{e}^{-t/T}\Bigg\{&u(0) - \Delta\left[v(0) - \frac{\chi w^0/T}{\chi^2+\Delta^2+1/T^2}\right]\frac{\sin\beta t}{\beta} \\
&+ \Delta\left[\Delta u(0) + \chi w(0) - \frac{\chi w^0/T^2}{\chi^2+\Delta^2+1/T^2}\right]\frac{\cos\beta t - 1}{\beta^2} + \frac{\Delta\chi w^0}{\chi^2+\Delta^2+1/T^2}\Bigg\} \\
&- \frac{\Delta\chi w^0}{\chi^2+\Delta^2+1/T^2}\,,
\end{aligned}
\tag{1.34a}
$$

$$
\begin{aligned}
v(t) = \mathrm{e}^{-t/T}\Bigg\{&\left[v(0) - \frac{\chi w^0/T}{\chi^2+\Delta^2+1/T^2}\right]\cos\beta t \\
&+ \left[\Delta u(0) + \chi w(0) - \frac{\chi w^0/T^2}{\chi^2+\Delta^2+1/T^2}\right]\frac{\sin\beta t}{\beta}\Bigg\} + \frac{\chi w^0/T}{\chi^2+\Delta^2+1/T^2}\,,
\end{aligned}
\tag{1.34b}
$$

$$
\begin{aligned}
w(t) = \mathrm{e}^{-t/T}\Bigg\{&w(0) - w^0 - \chi\left[v(0) - \frac{\chi w^0/T}{\chi^2+\Delta^2+1/T^2}\right]\frac{\sin\beta t}{\beta} \\
&+ \chi\left[\Delta u(0) + \chi w(0) - \frac{\chi w^0/T^2}{\chi^2+\Delta^2+1/T^2}\right]\frac{\cos\beta t - 1}{\beta^2} + \frac{\chi^2 w^0}{\chi^2+\Delta^2+1/T^2}\Bigg\} \\
&+ w^0\left(1 - \frac{\chi^2}{\chi^2+\Delta^2+1/T^2}\right).
\end{aligned}
\tag{1.34c}
$$

Here, the initial conditions $u(0)$, $v(0)$ and $w(0)$ at $t = 0$ are not assumed to be zero and we recall that $w^0 = \rho^0_{22} - \rho^0_{11}$ is the occupation probability difference in the absence of external radiation. The last term in eqs. (1.34a, b and c) is the steady-state value; it will be of interest in the case of steady-state preparation. The transient solutions will be of interest for the case of pulse preparation, as in an echo experiment.

Proceeding as before, we adopt the initial conditions (1.28) $B(0) = [0, 0, w(0)]$, and find that the Doppler-averaged density matrix element is

$$
\begin{aligned}
\langle\tilde{\rho}_{12}\rangle &= \frac{1}{ku\sqrt{\pi}}\int_{-\infty}^{\infty}\frac{1}{2}(u+iv)^{-(\Delta/ku)^2}\,\mathrm{d}\Delta \\
&\approx \frac{i\sqrt{\pi}}{2ku}\,\chi w(0)\,\mathrm{e}^{-(\Delta_1/ku)^2}\mathrm{e}^{-t/T}\Bigg\{J_0(\chi t) + \frac{2w^0/w(0)}{T\sqrt{\chi^2+1/T^2}}\,\mathrm{e}^{t/T} \\
&\quad - \frac{2w^0/w(0)}{\pi T}\int_0^{\infty}\frac{\mathrm{d}\Delta}{\chi^2+\Delta^2+1/T^2} \\
&\quad \times\left(\cos\sqrt{\Delta^2+\chi^2}\,t + \frac{\sin\sqrt{\Delta^2+\chi^2}\,t}{T\sqrt{\Delta^2+\chi^2}}\right)\Bigg\}\,.
\end{aligned}
\tag{1.35}
$$

The leading term of (1.35) is the same result as eq. (1.32) except that it is damped by the factor $\mathrm{e}^{-t/T}$. The second term is the steady-state value of $\langle\tilde{\rho}_{12}\rangle$ and for the present purpose can be ignored. The remaining two integrals, which cannot be evaluated analytically, can be estimated by replacing the trigonometric functions by unity; they are smaller than the leading term by $\sim 1/\chi T$ and $\sim 1/(\chi T)^2$ respectively; these terms can be neglected usually since $\chi T \sim 10^2$. The nutation signal field then becomes

$$
E_s(L,t) = \frac{2\pi^{3/2}}{u}(NL)\,\chi w(0)\,\mu_{12}\mathrm{e}^{-(\Delta_1/ku)^2}\,\mathrm{e}^{-t/T}J_0(\chi t)\cos(\Omega t - kL)\,.
\tag{1.36}
$$

1.5. Free induction decay

Let us assume that a molecular sample is resonantly excited under steady-state conditions by a laser beam and that suddenly the excitation is terminated. Under these conditions, the sample will radiate an intense coherent beam of light which has been called "optical free induction decay" [9] (FID) by analogy with the NMR effect first seen by Hahn [10]. For the present purpose we shall assume that the preparative stage ends by switching the molecular transition frequency out of resonance with the laser frequency. The switching mechanism is the Stark effect and will be discussed in the next section.

The preparative steady-state solutions for the period $t \leqslant 0$ can be derived from the Bloch equations (1.15) by setting the time derivatives equal to zero and yield

$$
u(0) = -\Delta\chi w^0/(\chi^2 T_1/T_2 + \Delta^2 + 1/T_2^2)\,,
\tag{1.37}
$$

$$v(0) = (\chi w^0/T_2)/(\chi^2 T_1/T_2 + \Delta^2 + 1/T_2^2), \tag{1.38}$$

$$w(0) = w^0[1 - (\chi^2 T_1/T_2)/(\chi^2 T_1/T_2 + \Delta^2 + 1/T_2^2)]. \tag{1.39}$$

These are the three time-independent terms of (1.34), but without the restriction that $T_1 = T_2$.

At time $t = 0$, we assume that the molecular transition frequency has been shifted by $\Delta\omega_{21}$ so that

$$\Delta \to \Delta' = \Delta + \Delta\omega_{21}, \tag{1.40}$$

and that the sample is sufficiently far out of resonance with laser light that we may set $\chi = 0$. The Bloch equations are then of the form

$$\dot{u} + \Delta' v + u/T_2 = 0, \tag{1.41a}$$

$$\dot{v} - \Delta' u + v/T_2 = 0, \tag{1.41b}$$

$$\dot{w} + (w - w^0)/T_1 = 0, \tag{1.41c}$$

and the solutions for $t > 0$ are given by

$$u(t) = [u(0)\cos\Delta' t - v(0)\sin\Delta' t]\,\mathrm{e}^{-t/T_2}, \tag{1.42a}$$

$$v(t) = [u(0)\sin\Delta' t + v(0)\cos\Delta' t]\,\mathrm{e}^{-t/T_2}, \tag{1.42b}$$

$$w(t) = w^0 + [w(0) - w^0]\,\mathrm{e}^{-t/T_1}. \tag{1.42c}$$

To obtain the field radiated, we again take the molecular velocity average expressed by (1.22)

$$\langle\tilde{\rho}_{12}\rangle = \frac{i}{2\sqrt{\pi}ku}\int_{-\infty}^{\infty} \mathrm{e}^{-(\Delta/ku)^2}\,[u(0)\sin\Delta' t + v(0)\cos\Delta' t]\,\mathrm{e}^{-t/T_2}\,\mathrm{d}\Delta,$$

$$\approx \frac{i}{2\sqrt{\pi}ku}\,\mathrm{e}^{-(\Delta_1/ku)^2}\chi w^0 \mathrm{e}^{-t/T_2}\cos\Delta\omega_{21}t$$

$$\times \int_{-\infty}^{\infty}\frac{-\Delta\sin\Delta t + (1/T_2)\cos\Delta t}{\chi^2 T_1/T_2 + \Delta^2 + 1/T_2^2}\,\mathrm{d}\Delta,$$

$$\approx \frac{i\sqrt{\pi}}{2ku}\,\mathrm{e}^{-(\Delta_1/ku)^2}\,\chi w^0 \exp[-t/T_2(1 + \sqrt{\chi^2 T_1 T_2 + 1})]$$

$$\times\left(\frac{1}{\sqrt{\chi^2 T_1 T_2 + 1}} - 1\right)\cos\Delta\omega_{21}t. \tag{1.43}$$

Here, $\langle u\rangle = 0$ and we have assumed that the Doppler factor can be taken outside the integral. The field amplitude (1.24) associated with the induced polarization is

$$E_{12} = -i(2\pi\Omega/c)\,NL\mu_{12}\langle\tilde{\rho}_{12}\rangle, \tag{1.44}$$

whereas the total field, including that of the laser, is

$$E_{\mathrm{T}} = (E_{12} + \tfrac{1}{2}E_0)\,\mathrm{e}^{i(\Omega t - kz)} + \mathrm{c.c.} \tag{1.45}$$

The intensity, therefore, contains a cross term or beat

$$(E^2)_{\mathrm{beat}} = 2E_{12}E_0$$

$$= E_0 Q_{12}(t)\cos\Delta\omega_{12}t, \tag{1.46a}$$

where

$$Q_{12}(t) = 2\pi^{3/2}NL\mu_{12}^2 E_0 w^0\left(\frac{1}{\sqrt{\chi^2 T_1 T_2 + 1}} - 1\right)$$

$$\times \exp[-(\Delta_1/ku)^2]\exp[-t/T_2(1 + \sqrt{\chi^2 T_1 T_2 + 1})]. \tag{1.46b}$$

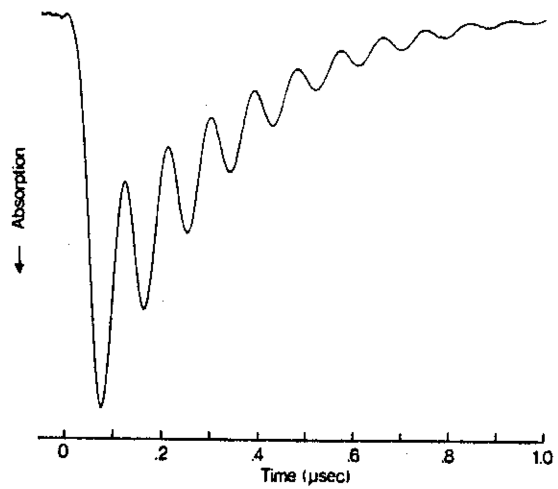


Fig. 3. Optical free induction decay in NH_2D following a step function Stark field. The beat frequency is the Stark shift, and the slowly varying background is a nutation signal of a second velocity group, more clearly shown in fig. 2. (From Brewer and Shoemaker [9].).

The decay behavior of (1.46) has two contributions [8,9]: (i) a homogeneous part with time constant T_2 and (ii) an inhomogeneous part with time constant $T_2/\sqrt{\chi^2 T_1 T_2 + 1}$ that reflects the velocity bandwidth excited during steady-state preparation. At moderately high laser intensities (a few W/cm^2), the inhomogeneous dephasing can be dominant, the free induction signal will then decay rapidly and appear modulated at a frequency given by the Stark shift $\Delta\omega_{21}$. The subtle behavior near the time origin is discussed elsewhere. An experimental demonstration of the FID effect is shown in fig. 3.

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Vector Model

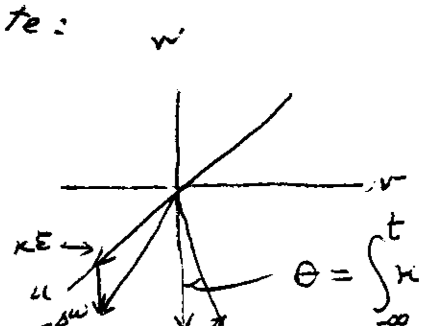
The vector model for the pseudopolarization can be defined in several different ways depending upon the definition of the pseudopol. $\vec{P}$ and the angular frequency or effective field Ω. I prefer the following which places $\vec{P}$ pointing down for an atom in the ground state:

$$\vec{P} \equiv u\hat{u}_0 + v\hat{v}_0 + w\hat{w}_0$$

$$\vec{\Omega} = \kappa\mathcal{E}\,\hat{u}_0 - \Delta\omega\,\hat{w}_0$$

$$\Delta\omega = \omega_0 - \omega_{laser}$$

$$\dot{\vec{P}} = \vec{\Omega}\times\vec{P} = \begin{vmatrix} \hat{u}_0 & \hat{v}_0 & \hat{w}_0 \\ \kappa\mathcal{E} & 0 & -\Delta\omega \\ u & v & w \end{vmatrix}$$



$$\theta = \int_{-\infty}^{t} \kappa\,\mathcal{E}(t)\,dt$$

$$\dot{u} = v\Delta\omega \qquad - u/T_2'$$

$$\dot{v} = -u\Delta\omega - \kappa\mathcal{E}w \qquad - v/T_2'$$

$$\dot{w} = \kappa\mathcal{E}v \qquad - (w - w_0)/T_1$$

$$\kappa = 2p/\hbar$$

$$\frac{1}{\tau_{21}} = \frac{4}{3}\frac{\omega^3}{\hbar c^3}(\sqrt{2}p)^2 \qquad p = p_{MH} = p_{Schiff}/\sqrt{2}$$

Rb $\tau_{21} = 42$ ns

$\lambda = 7947$ Å

$\Rightarrow p = 4.35\times10^{-18}$ esu-cm

$$\kappa\mathcal{E}\tau_p = \pi \qquad \pi \text{ pulse}$$

$$I = \frac{c\mathcal{E}^2}{4\pi} \xrightarrow[\tau_p = 5\,ns]{} 1.4\ W/cm^2$$

$s_i(t) \equiv \langle \hat{\sigma}_i(t) \rangle$

If 2 and 1 are connected by $\Delta m = 0$, then select phases s.t. $\vec{d}_i = 0$ and set $\frac{2}{\hbar}\vec{d}_r \cdot \vec{E} = \kappa E$, $\kappa \equiv 2d/\hbar$ $\Rightarrow$

$$\begin{cases} \dot{s}_1(t) = -\omega_0 s_2(t) \\ \dot{s}_2(t) = \omega_0 s_1(t) + \kappa E(t, \vec{r}_0) s_3(t) \\ \dot{s}_3(t) = -\kappa E(t, \vec{r}_0) s_2(t) \end{cases}$$

$s_1 = a^*b + ab^*$
$s_2 = -i(a^*b - ab^*)$
$s_3 = |a|^2 - |b|^2$
$\psi = a|2\rangle + b|1\rangle$

$$\Rightarrow \frac{d\vec{s}(t)}{dt} = \vec{\Omega}^F(t) \times \vec{s}(t)$$

$$\vec{\Omega}^F = (-\kappa E, 0, \omega_0)$$

$\kappa E \ll \omega_0$; if $\kappa E \approx \omega_0$ $\Rightarrow$ $E \approx 1V/a_0 \approx 10^9$ V/cm $\quad 10^{15}$ W/cm^2
$\approx$ field ionization energy

<u>Rotating Wave Approximation</u>

$$E(t) = \mathcal{E}(t)[e^{i\omega t} + c.c.] = 2\mathcal{E}(t)\cos\omega t$$

$$\vec{\Omega}^F = \underbrace{(0, 0, \omega_0)}_{\vec{\Omega}^0} + \underbrace{(-\kappa\mathcal{E}\cos\omega t, -\kappa\mathcal{E}\sin\omega t, 0)}_{\vec{\Omega}^+} + \underbrace{(-\kappa\mathcal{E}\cos\omega t, +\kappa\mathcal{E}\sin\omega t, 0)}_{\vec{\Omega}^-}$$

$\omega_0 \approx \omega$ $\quad$ $\omega \approx \omega_0$ $\quad$ $= -\omega$

$\approx$ const. in rotating frame

counter-rotating at 2ω
— RWA ignore

$$\Rightarrow \quad \dot{s}_1 = -\omega_0 s_2 - \kappa\mathcal{E} s_3 \sin\omega t$$
$$\dot{s}_2 = \omega_0 s_1 + \kappa\mathcal{E} s_3 \cos\omega t$$
$$\dot{s}_3 = -\kappa\mathcal{E}[s_2\cos\omega t - s_1 \sin\omega t]$$

Define a nearly stationary vector ρ in the rotating frame

$$\begin{pmatrix} u \\ v \\ w \end{pmatrix} = \begin{pmatrix} \cos\omega t & \sin\omega t & 0 \\ -\sin\omega t & \cos\omega t & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} s_1 \\ s_2 \\ s_3 \end{pmatrix} \Rightarrow \begin{aligned} \dot{u} &= -(\omega_0 - \omega) v \\ \dot{v} &= (\omega_0 - \omega) u + \kappa\mathcal{E} w \\ \dot{w} &= -v\kappa\mathcal{E} \end{aligned}$$

$$\Rightarrow \frac{d\vec{\rho}}{dt} = \vec{\Omega} \times \vec{\rho}$$
$$\vec{\Omega} = (-\kappa\mathcal{E}, 0, \omega_0 - \omega)$$

$\frac{\hbar\omega_0}{2} w$ expectation of the atom's unperturbed energy

u and $-v$ are the components, in units of the transition moment d, of the atomic dipole moment in-phase and in-quadrature with E.

$\dot{w} \sim v\mathcal{E}$ $\Rightarrow$ v produces energy changes — absorptive component

OPTICAL BLOCH EQNS.

Allen + Eberly approach:

$$\hat{\mathcal{H}} = \hat{H}_A - \hat{d} \cdot \hat{\vec{E}}(\vec{r}_0)$$

electric field operator evaluated at the position $\vec{r}_0$ of the dipole

dipole moment operator

$\langle 2|\hat{d}|1\rangle = d_{21}$ $\quad \langle 1|\hat{d}|2\rangle = d_{21}^*$

$\vec{d}_{21} = \vec{d}_r + i\vec{d}_i$
real imag.

$$\hat{d} \Rightarrow \begin{pmatrix} 0 & \vec{d}_r + i\vec{d}_i \\ \vec{d}_r - i\vec{d}_i & 0 \end{pmatrix} = \vec{d}_r\hat{\sigma}_1 - \vec{d}_i\hat{\sigma}_2$$

$[\hat{\sigma}_1, \hat{\sigma}_2] = 2i\hat{\sigma}_3 \quad \ldots$

Heisenberg method

$i\hbar\dot{\hat{O}} = [\hat{O}, \hat{\mathcal{H}}]$ for any operator $\hat{O}$ not explicitly time-dep.

$$\hat{\mathcal{H}} = \tfrac{1}{2}(W_+ + W_-)\hat{I} + \tfrac{1}{2}(W_+ - W_-)\hat{\sigma}_3 - (\vec{d}_r\cdot\hat{\vec{E}})\hat{\sigma}_1 + (\vec{d}_i\cdot\hat{\vec{E}})\hat{\sigma}_2$$

$$\dot{\hat{\sigma}}_1 = -\omega_0\hat{\sigma}_2(t) + \frac{2}{\hbar}[\vec{d}_i\cdot\hat{\vec{E}}(t)]\hat{\sigma}_3(t)$$
$$\dot{\hat{\sigma}}_2 = \omega_0\hat{\sigma}_1(t) + \frac{2}{\hbar}[\vec{d}_r\cdot\hat{\vec{E}}(t)]\hat{\sigma}_3(t) \qquad \omega_0 = \frac{W_+ - W_-}{\hbar}$$
$$\dot{\hat{\sigma}}_3 = -\frac{2}{\hbar}[\vec{d}_r\cdot\hat{E}(t)]\hat{\sigma}_2(t) - \frac{2}{\hbar}[\vec{d}_i\cdot\hat{E}(t)]\hat{\sigma}_1(t)$$

Complex eqns. "No very general solutions are known."

If quantum correlations can be ignored, then operator products such as $\hat{\vec{E}}(t)\hat{\sigma}_3(t)$ can be factored in every expectation value

$$\langle\hat{E}(t)\hat{\sigma}_3(t)\rangle = \langle\hat{E}(t)\rangle\langle\hat{\sigma}_3(t)\rangle.$$

"We define the semiclassical radiation theory of two-level atoms to be the theory resulting from a consistent application of such a factorization to the $\hat{\sigma}$ eqns. above." The semiclassical theory uses the expectation value rather than the operator Maxwell eqs.. $\langle\hat{\vec{E}}(t, \vec{r}_0)\rangle$ is interpreted to be a purely classical electric field. SC theory is not general enough to describe spontaneous emission correctly.

Allen + Eberly 34–38

Bloch-Siegert Shift

RWA can give rise to a shift in the true resonance frequency

$\vec{\Omega} = (-2\kappa\mathcal{E}\cos\omega t,\ 0,\ \omega_0)$ in lab. frame

$\vec{\Omega}(+\omega) = ?$ in frame rotating right about the 3 axis at ω?

$$\vec{\Omega}(+\omega) = \begin{pmatrix} \cos\omega t & \sin\omega t & 0 \\ -\sin\omega t & \cos\omega t & 0 \\ 0 & 0 & 1 \end{pmatrix}\begin{pmatrix} -2\kappa\mathcal{E}\cos\omega t \\ 0 \\ \omega_0 \end{pmatrix} - \begin{pmatrix} 0 \\ 0 \\ \omega \end{pmatrix}$$

$$= \begin{pmatrix} -2\kappa\mathcal{E}\cos^2\omega t \\ 2\kappa\mathcal{E}\cos\omega t\sin\omega t \\ \omega_0 - \omega \end{pmatrix} = \begin{pmatrix} -\kappa\mathcal{E}(1+\cos 2\omega t) \\ \kappa\mathcal{E}\sin 2\omega t \\ \omega_0 - \omega \end{pmatrix}$$

Is there a frame in which the natural precession + the rotating part of the torque have not only the same freq., but also the same dir. of rot.?

counter-rotating with $\omega \approx -\omega_0 \Rightarrow$ freq. of rotating part 2ω $\approx$ nat. precess. freq. $\omega_0 - \omega \approx 2\omega$

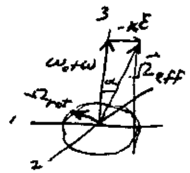
$\Omega(-\omega) = [-\kappa\mathcal{E}(1+\cos 2\omega t),\ -\kappa\mathcal{E}\sin 2\omega t,\ \omega_0+\omega] = \vec{\Omega}_{eff} + \vec{\Omega}_{rot}$

$\vec{\Omega}_{rot} = -\hat{1}\,\kappa\mathcal{E}\cos 2\omega t - \hat{2}\,\kappa\mathcal{E}\sin 2\omega t$

static "effective" torque $\vec{\Omega}_{eff}$ is simply the constant part of $\vec{\Omega}(-\omega)$

$\vec{\Omega}_{eff} = \hat{u}_{eff}\sqrt{\kappa^2\mathcal{E}^2 + (\omega+\omega_0)^2}$

$\hat{u}_{eff} = -\hat{1}\sin\alpha + \hat{3}\cos\alpha$



new coordinate axes:

$\hat{2}' = \hat{2}$, $\hat{3}' = \hat{u}_{eff}$ $\quad \hat{1}' = \hat{1}\cos\alpha + \hat{3}\sin\alpha$ $\quad \hat{3}'\cos\alpha + \hat{1}'\sin\alpha = \hat{3}$, $\ \hat{1}'\cos\alpha - \hat{3}'\sin\alpha = \hat{1}$

$$\vec{\Omega}_{rot} = -\kappa\mathcal{E}\left[\cos 2\omega t\,(\hat{1}'\cos\alpha - \hat{3}'\sin\alpha) + \sin 2\omega t\,(\hat{2}')\right]$$

$$= \kappa\mathcal{E}\sin\alpha\cos 2\omega t\ \hat{3}'$$

$$\left.\begin{aligned} &-\kappa\mathcal{E}\left(\tfrac{1+\cos\alpha}{2}\right)[\hat{1}'\cos 2\omega t + \hat{2}'\sin 2\omega t] \\ &+\kappa\mathcal{E}\left(\tfrac{1-\cos\alpha}{2}\right)[\hat{1}'\cos 2\omega t - \hat{2}'\sin 2\omega t] \end{aligned}\right\}$$ driving torques $\perp$ static Ω_{eff} + rotating about it at $\pm 2\omega$

$\Rightarrow$ two resonance conditions $|\Omega_{eff}| = \sqrt{(\kappa\mathcal{E})^2 + (\omega+\omega_0)^2} = \pm 2\omega$

Allen + Eberly 47-50

$$4\omega^2 = \kappa^2\mathcal{E}^2 + (\omega_0+\omega)^2$$

$$\omega^2 - \frac{2\omega_0}{3}\omega - \frac{\kappa^2\mathcal{E}^2+\omega_0^2}{3} = 0$$

$$\omega = \left(\frac{2\omega_0}{3} \pm \sqrt{\frac{4}{9}\omega_0^2 + \frac{4}{3}(\kappa^2\mathcal{E}^2+\omega_0^2)}\right)\Big/2 = \frac{\omega_0}{3} \pm \frac{2\omega_0}{3}\left[1 + \frac{3}{4}\left(\frac{\kappa\mathcal{E}}{\omega_0}\right)^2\right]^{1/2}$$

$$\approx \frac{\omega_0}{3}\left\{1 \pm 2\left[1 + \frac{3}{8}\left(\frac{\kappa\mathcal{E}}{\omega_0}\right)^2\right]\right\}$$

$$\frac{1-\cos\alpha}{1+\cos\alpha} \approx \left(\frac{\alpha}{2}\right)^2 \approx \frac{1}{4}\left(\frac{\kappa\mathcal{E}}{\omega_0}\right)^2 \ll 1 \quad\Rightarrow\quad \omega = \omega_0\left[1 + \frac{1}{4}\left(\frac{\kappa\mathcal{E}}{\omega_0}\right)^2 + \cdots\right]$$

$$\delta\omega_{B\text{-}S} = \frac{1}{4}\left(\frac{\kappa\mathcal{E}}{\omega_0}\right)^2$$

1ns, π-pulse $\quad \kappa\mathcal{E}\tau_p \approx \pi$

$\kappa\mathcal{E} \approx \pi/\tau_p \approx \pi\times 10^9$

$$\frac{\delta\omega_{B\text{-}S}}{\omega_0} \approx \frac{1}{4}\left(\frac{\kappa\mathcal{E}}{\omega_0}\right)^2 \approx \frac{1}{4}\left(\frac{\cancel{\pi}\times 10^9\,s^{-1}\ \cancel{6}\times 10^{-5}\,cm}{\cancel{2\pi}\ \cancel{3}\times 10^{10}\,cm\,s^{-1}}\right)^2$$

$$\approx \frac{10^{-12}}{4}$$

$\delta\nu_{B\text{-}S} \approx 0.5\times 10^{15}\,\frac{10^{-12}}{4} \approx \underline{\underline{100\ Hz}}$ very small

Allen + Eberly 50

Circularly polarized light — optical Bloch equations are exact — no Bloch-Siegert shift

B-S shift comes from LP $\updownarrow$ = ↷ + ↶

CP = ↷ (or ↶)

So in rotating frame $\mathcal{E}$ is stationary — no counter rotating component.

Most of our careful self induced transparency expts. were with CP — but since $\delta\nu_{B\text{-}S}$ is so small it doesn't matter anyhow.

Allen + Eberly 107–109

Optical Nutation

see Brewer pp. 347–351 (73)–(75)

$$P(z,t) = \frac{u + iv}{2} N\mu_{12}\, e^{i(\Omega t - kz)} + c.c.$$

u in phase comp.

v out of phase by 90°

$$E_x(z,t) = E_0 \cos(\Omega t - kz)$$

$$\frac{u+iv}{2} = \langle \tilde\rho_{12} \rangle$$

nutating atoms radiate a signal field

$$E_s(z,t) = E_{12}(z,t)\, e^{i(\Omega t - kz)} + c.c.$$

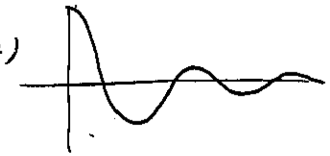
$$\frac{\partial E_{12}}{\partial z} = -2\pi i k N \mu_{12} \langle \tilde\rho_{12} \rangle$$

$$\cdots\quad E_s(L,t) = \frac{2\pi^{3/2}}{u}(NL)\,\chi w(0)\mu_{12}\, e^{-(\Delta_1/ku)^2} J_0(\chi t) \underbrace{\cos(\Omega t - kL)}_{\text{carrier oscillation}}$$

on resonance so $\langle u \rangle \sim 0$ $\quad$ u rms vel.

bandwidth excited assumed << Doppler width ku

$J_0(\chi t)$

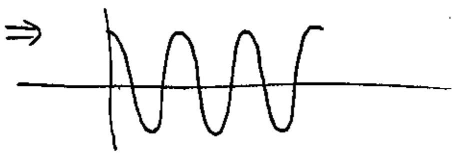


comes from $\int_{-\infty}^{\infty} \left(\frac{u+iv}{2}\right) e^{-(\Delta/ku)^2} d\Delta$

$$\longrightarrow e^{-(\Delta_1/ku)^2} \int_0^\infty \frac{\sin\sqrt{\Delta^2 + \chi^2}\, t}{\sqrt{\Delta^2+\chi^2}}\, d\Delta$$

no Doppler broadening $\Rightarrow$ no $\int d\Delta$

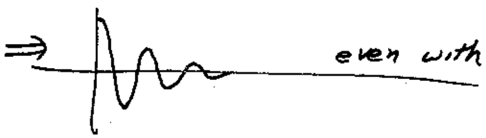
$\Rightarrow$



"forever"

Realistically — always damping $\Rightarrow e^{-t/T}$ factor

$\Rightarrow$



even with no Doppler broadening

See p. (77) Rb π pulse $I = 1.4\ \mathrm{W/cm^2}$ for 5 ns pulse

$\kappa \mathcal{E} \tau_p = \pi \qquad \kappa\mathcal{E} = \frac{\pi}{\tau_0} \qquad \frac{\kappa\mathcal{E}}{2\pi} = \nu_{Rabi} = \frac{1}{2\tau_0} = 100\ \mathrm{MHz}$

FREE INDUCTION DECAY

$$\dot u = -\Delta v - u/T_2'$$

$$\dot v = \Delta u - v/T_2' + \kappa\mathcal{E} w$$

$$\dot w = -\frac{(w - w_{eq})}{T_1} - \kappa \mathcal{E} v$$

$\Delta = 0 \quad u = 0$

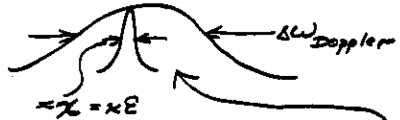
$\bar v = \kappa \mathcal{E} w T_2'$ and v decays as $e^{-t/T_2'}$ — no dephasing

distribution of Δ's

Brewer, p. 352 $\quad \int v(\Delta\omega)\, e^{-(\Delta\omega/ku)^2} e^{-t/T_1}\, d\omega$

$$\cdots \quad e^{-\frac{t}{T_2}\left(1 + \sqrt{\chi^2 T_1 T_2 + 1}\right)}$$

enhanced decay from dephasing by frequency band excited:



$v(0) = ?\quad \Delta(-\Delta v T_2') - v/T_2' + \kappa\mathcal{E}\,(w_0/T_1 - \kappa\mathcal{E} v)\,T_1 = 0$

$$v(0) = \frac{\kappa\mathcal{E} w_0 / T_2'}{\frac{\kappa^2\mathcal{E}^2 T_1}{T_2'} + \Delta^2 + \frac{1}{(T_2')^2}} \quad \text{so width} \quad \sim \frac{\kappa\mathcal{E} T_1^{1/2}}{(T_2')^{1/2}}$$

$$\frac{\sqrt{\chi^2 T_1 T_2}}{T_2} = \chi\sqrt{\frac{T_1}{T_2}} \longrightarrow \kappa\mathcal{E}\sqrt{\frac{T_1}{T_2'}}$$

For ex.

$T_2' = 40\ \mathrm{ns} = T_1$

$\chi = \kappa\mathcal{E} = \omega_{Rabi} \approx (100\ \mathrm{MHz})\,2\pi$

$\sqrt{\chi^2 T_1 T_2} = 2\pi\, 10^8\, \mathrm{s^{-1}}\; 40\times 10^{-9}\, \mathrm{s} = 8\pi >> 1$

With ps pulses the entire Doppler width can be excited, then $e^{-t\frac{1}{T_2^*}}$ where $\frac{1}{\pi T_2^*} \approx \Delta\nu_{Dop.}$

$\Delta\nu \approx 3\ \mathrm{GHz} \qquad T_2^* \approx 0.1\ \mathrm{ns}$

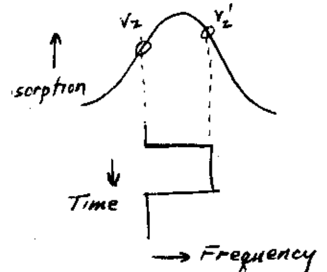
Optical Coherent Transients by Laser Frequency Switching

A. Z. Genack & R. G. Brewer, Phys. Rev. A 17, 1463 (1978).
R. G. Brewer & A. Z. Genack, Phys. Rev. Lett. 36, 959 (1976).

time scale — ms to ~50 ps

The frequency of a cw dye laser is switched with an intracavity electro-optic modulator.

The dye's dephasing time of a ≤ few ps is too rapid to interfere. The method incorporates all of the advantages inherent in Stark switching, including heterodyne detection & high sensitivity, without being restricted to Stark-tunable systems.

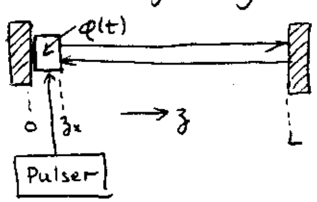


Heterodyne detection occurs automatically because the coherent light radiated by the sample overlaps the laser beam in space & time & the two are displaced in frequency due to the laser freq. shift. Thus, the sample retains memory of the initial laser freq. in the preparative stage whereas the laser at its new freq. acts as a local oscillator.

Standing-wave optical field in a cavity of optical length L:

$$E(z,t) = E_0\left[e^{i(\omega t - kz)} + e^{i(\omega t + kz)}\right] + c.c.$$

$\omega = 2\pi\nu/T$ ν large integer $T = 2L/c$ round-trip time



$\varphi(t)$ produced by phase modulator

$z_x \ll L$

consider one running wave

$$E(z,t) = E_0\, e^{i(\omega t - kz)}$$

$$\phi = k z_x$$

$$\phi(t) = \dot\phi t \qquad 0 < t < t_1$$
$$= \dot\phi t_1 \qquad t_1 < t$$
$$\dot\phi = \text{const.}$$

next to one mirror ⇒ double pass

$$\phi(t) = \dot\phi t = 2\frac{\omega}{c} z_x \dot n t$$

$\dot n$: modulator's refractive index time derivative

$$E(z_x, t) = E_0\, e^{i(\omega - \dot\phi)t - ikz_x} \qquad 0 < t < t_1$$
$$= E_0\, e^{i[\omega t - \phi(t_1)] - ikz_x} \qquad t_1 < t < T'$$

where the new cavity round-trip time is T'.

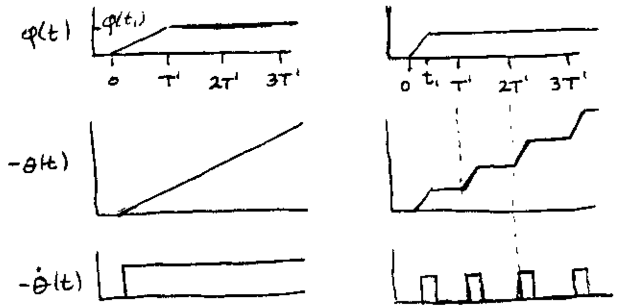
The freq. shift $\dot\phi = 2\omega z_x \dot n/c$ takes the form of a Doppler shift if we associate $z_x \dot n$ with the effective translation velocity of the modulator.

$t > t_1$, the phase change $\phi(t_1)$ implies the additional retardation $\Delta T = \phi(t_1)/\omega = 2 z_x \dot n t/c$ each time the wave train passes through the modulator twice. Thus, the light wave circulates indefinitely at the new round-trip time $T' = T + \Delta T$.

$\varphi(t) \equiv$ phase modulation as a fnc. of time

$\theta(t) \equiv$ resulting phase of the light wave

$\dot\theta(t) =$ its time derivative which illustrates the instantaneous freq.



before switching —
$$E(t) = E(t-T), \quad t < 0$$
after switching —
$$E(t) = E(t-T)e^{-i\phi(t)}, \quad t > 0$$
where the round-trip time changes from $T \to T'$ due to the phase factor $e^{-i\phi(t)}$

$$E(t) = E_0 e^{i[\omega t + \theta(t)]}$$

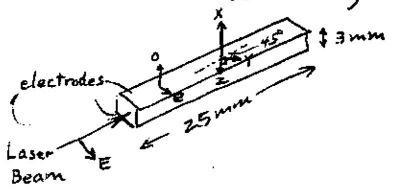
$$E_0 e^{i[\omega(t-T) + \theta(t-T)] - i\phi(t)} = E_0 e^{i[\omega t + \theta(t)]}$$

$$\Rightarrow \boxed{\theta(t) = \theta(t-T) - \phi(t)} \quad \text{since } e^{-i\omega T} = e^{-i2\pi\nu} = 1$$

For a phase shifter <u>outside</u> the cavity, a frequency shift occurs only when φ is changing, so if long pulses are needed then one must change φ over a long time $\Rightarrow$ high voltage ramp

Experimental: electro-optic phase modulator

AD*P crystal



0.6 MHz/V

maximum shift = 300 MHz

(laser's axial mode spacing = 390 MHz)

Important: There is no slow buildup — the frequency shift is instantaneous. Only the modulator transit time $\frac{\ell}{c}n \approx 50ps$

<u>FID</u>

inhomogeneous dephasing time T_2^* plus homogeneous relaxation

<u>Optical Nutation</u>

$$e^{-\frac{1}{2}\left(\frac{1}{\tau_1}+\frac{1}{\tau_2}\right)t} \underbrace{J_0(Xt)}_{\text{dephasing}}$$

$$\frac{1}{\tau_1} = \frac{1}{T_1}$$

$$\frac{1}{\tau_2} = \frac{1}{T_2'}$$

<u>Photon Echo</u> transverse relax. with inhomogeneous (reversible) part removed $\Rightarrow T_2'$

<u>Two Pulse Opt. Nut.</u> $S_\infty - S(t=\tau) = e^{-\gamma_a t}\left(1+\frac{\gamma_1}{\gamma_a-\gamma_b}\right)+e^{-\gamma_b t}\left(1-\frac{\gamma_1}{\gamma_a-\gamma_b}\right)$

$$\begin{array}{ll} a \;\text{———}\; \gamma_a & \\ \quad \gamma_1 \downarrow & \\ b \;\text{———}\; \gamma_b & \end{array}$$

POINT BEING MADE — with several optical coherent transient experiments all of the relaxation times can be determined

Another good reference —

R. G. Brewer, "Coherent Optical Transients," Physics Today <u>30</u>, No. 5, May, 50 (1977).

<u>Advantage of heterodyne</u> for small signal E_s

$$E \sim E_0 \cos\omega t + E_s \cos\omega_s t$$

$$I \sim \overline{E^2} \sim \frac{E_0^2}{2} + \frac{E_s^2}{2} + 2E_0E_s\overline{\cos\omega t\cos\omega_s t}$$

$$= \frac{E_0^2}{2} + \frac{E_s^2}{2} + E_0E_s\left[\overset{\to 0}{\cancel{\cos(\omega+\omega_s)t}} + \cos(\omega-\omega_s)t\right]$$

If $E_s \ll E_0$ then $E_0E_s \gg E_s^2$ $\Rightarrow$ enhanced sensitivity.

Heterodyne signal oscillates at $\omega-\omega_s$ $\Rightarrow$ further discrimination against E_0^2

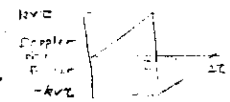
$\boxed{\text{PHOTON ECHOES}}$

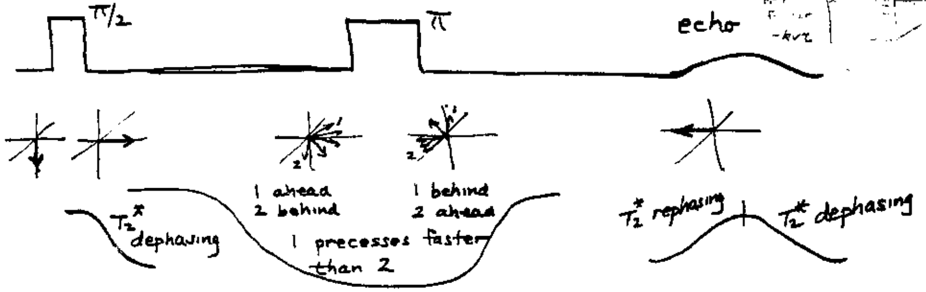
First observation:

N. A. Kurnit, I. D. Abella, S. R. Hartmann, Phys. Rev. Lett. <u>13</u>, 567 ('64),

I. D. Abella, N. A. Kurnit, S. R. Hartmann, Phys. Rev. <u>141</u>, 391 ('66).

Inasmuch as the phenomenon can be treated completely without appeal to the quantum theory of radiation, it might better be called "optical spin echo"





Echo detection depends upon free induction dephasing between the two excitation pulses and rephasing after the π pulse. So the echo works best in an inhomogeneously broadened system in which rapid reversible dephasing occurs

Sargent, Scully, Lamb, p. 216

via the velocity distribution (Doppler effect) or inhomogeneities in external electric or magnetic fields.

Simple derivation of PE

$$\Psi(\vec{r},t) = C_a(t)\, e^{-i\omega_a t} u_a(\vec{r}) + C_b(t)\, e^{-i\omega_b t} u_b(\vec{r}) \qquad \text{—— a}$$

$$\Psi(\vec{r},-0) = u_b(\vec{r}) \qquad \text{—— b}$$

$$\Psi(\vec{r},0) = \frac{1}{\sqrt{2}}(u_a + u_b) \quad \text{just after } 90^\circ \text{ or } \frac{\pi}{2} \text{ pulse}$$

$$\Psi(\vec{r},\tau_-) = \frac{1}{\sqrt{2}}\left(e^{-i\omega_a\tau} u_a + e^{-i\omega_b\tau} u_b\right) \quad \text{just before } \pi \text{ pulse}$$

$$\Psi(\vec{r},\tau) = \frac{1}{\sqrt{2}}\left(e^{-i\omega_b\tau} u_a + e^{-i\omega_a\tau} u_b\right) \quad \text{" after " "}$$

$$\Psi(\vec{r},\tau+\tau') = \frac{1}{\sqrt{2}}\left[e^{-i\omega_b\tau} e^{-i\omega_a\tau'} u_a + e^{-i\omega_a\tau} e^{-i\omega_b\tau'} u_b\right]$$

Polarization: $\langle e\vec{r}\rangle = \int d\omega\, W(\omega) \int d^3r\, |\Psi(\vec{r},\tau+\tau')|^2\, e\vec{r}$

$$= \frac{p}{2}\int d\omega\, W(\omega)\, e^{-i\omega(\tau'-\tau)} + c.c. \qquad \text{Clearly peaks for } \tau' = \tau$$

since $\langle u_a | \vec{r} | u_a \rangle = \langle u_b | \vec{r} | u_b \rangle = 0$

$\omega = \omega_a - \omega_b$

For a Gaussian frequency distribution

$$W(\omega) = \frac{1}{\sqrt{\pi}\,\Delta\omega}\, e^{-(\omega-\omega_0)^2/(\Delta\omega)^2} \qquad \Delta\omega = 2/T_2^*$$

$$\langle er\rangle = p\, e^{-(\tau'-\tau)^2(\Delta\omega)^2} \cos(\omega_0 t) \qquad \text{peaks at } \tau' = \tau$$

Intensity of the echo depends upon how many atomic polarizations rephase. If irreversible processes, such as spontaneous decay or a phase interrupting collision, occur they will diminish the echo intensity. Hence I_{echo} versus the time between pulses permits a determination of the dephasing time T_2'.

SSL 217–218

So long as the π pulse is symmetric, the echo (shape, magnitude, etc.) is independent of any aspect of the π pulse.

Directional character:

In Brewer's frequency shifting experiments, the echo is observed by switching into resonance twice for the $\pi/2 - \pi$ sequence and then observing the echo as a heterodyne signal beating with the cw oscillator.

In a two-pulse PE experiment there is often difficulty in separating the echo from the $\pi/2$ + π pulses since if they are collinear they give rise to an echo in the same direction. If the sample is optically thin to avoid propagation effects, the echo intensity is 10 to 100 times smaller than the excitation pulses. Relaxation, imperfect spatial overlap of " " ", etc. reduce the echo further. If the " " are close together, the detector may be unable to recover from the intense excitation pulses in time to detect an echo 10^{-3} to 10^{-5} times smaller than the π pulse. If the excitation pulses are separated by an angle ϕ, then the echo + π pulses " " " " " ϕ, also, permitting angular discrimination against $I_{\pi/2}$ + I_π.

Phase relations between dipoles are automatically satisfied for spin echoes because magnetic resonance wavelengths are comparable to, or even much larger than, the size of the radiating sample. For photon echoes with $L \gg \lambda$ the relative phasing is governed by the finite speed of light. For the ℓ^{th} atom

$$t_1 \longrightarrow t_{1\ell} = t_{10} + \hat{n}_1 \cdot \vec{r}_\ell / c \qquad \text{retarded time}$$

where t_{10} is the time required for the first ($\pi/2$) pulse to reach a fictional reference atom located at the origin of the coordinate system.

Allen + Eberly Chap. 9

$$P(t) = \mathcal{N} d \int d\Delta' g(\Delta') \frac{1}{N} \sum_{\ell=1}^{N} Re\left[\{u_\ell(t;\Delta') + i v_\ell(t;\Delta')\} e^{i\omega t}\right]$$

from simple approach on p. 88

$$p \sim e^{i\omega(\tau' - \tau)}$$

τ' = echo time, τ = π pulse time

but using retarded times

$$e^{i\omega(\tau'-\tau)} \longrightarrow e^{i\Delta'[(t_{40}-t_{30})-(t_{20}-t_{10})]}$$

$$\times\, e^{i\frac{\omega}{c}[(\hat{n}-\hat{n}_2)-(\hat{n}_2-\hat{n}_1)]\cdot\vec{r}_\ell}$$

$$t_{1\ell} = t_{10} + \hat{n}_1 \cdot \vec{r}_\ell/c$$
$$t_{2\ell} = t_{20} + \hat{n}_2 \cdot \vec{r}_\ell/c$$
$$t_{3\ell} = t_{30} + \hat{n}_2 \cdot \vec{r}_\ell/c$$
$$t_{4\ell} = t_{40} + \hat{n} \cdot \vec{r}_\ell/c$$

Sharply peaked for $\hat{n} + \hat{n}_1 - 2\hat{n}_2 = 0$

$\sin\phi = \sin\phi' \qquad \phi = \phi'$

$\cos\phi + \cos\phi' = 2 \approx (1 + \frac{\phi^2}{2} + \cdots) + (1 + \frac{\phi'^2}{2} + \cdots) = 2 + O(\phi^2)$

So to order ϕ^2, $\phi' = \phi$, i.e. $\angle_{echo-\pi} = \angle_{\pi-\pi/2}$.

How large can ϕ be? For max. echo, for all values of $\vec{r}_\ell$ from one end of the sample to the other, the exponent must change by much less than $2\pi i$ $\Rightarrow$ $\frac{\omega}{c}(2\hat{n}_2 - \hat{n}_1 - \hat{n})\cdot\vec{r}_\ell \ll 2\pi$

assume sample length is L + its major axis lies along $\hat{n}_2$

$$\Rightarrow \quad (2\hat{n}_2 - \hat{n}_1 - \hat{n})\cdot\hat{n}_2 L \ll \frac{2\pi c}{\omega} = \lambda$$

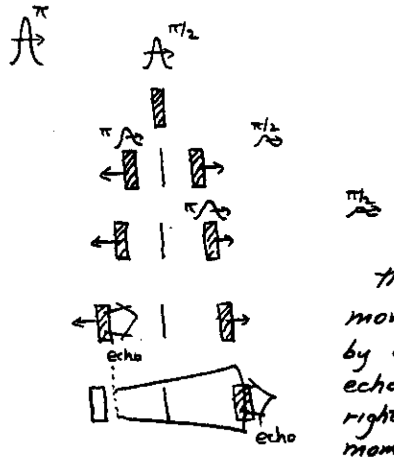
$$2 - \hat{n}_1\cdot\hat{n}_2 - \hat{n}\cdot\hat{n}_2 \ll \lambda/L$$

$$2(1-\cos\phi) \approx \boxed{\phi^2 \ll \lambda/L}$$

Above assumes no motion of atoms between pulses. In a gas $\Delta r \approx v\tau \approx (10^5 \frac{cm}{s}) 10^{-8} s \approx 10\mu m \gg \lambda$.

But, the dephasing due to atomic motion is exactly compensated in the course of the echo development.

Allen + Eberly 207–209



the echo emission from the left-moving atoms occurs earlier in time by the right amount so that their echo pulse just catches up with the right-moving atoms at exactly the moment *their* Bloch vectors rephase + emit their echo pulse.

A + E 210–211

Other references:

I. D. Abella, "Echoes at Optical Frequencies," in E. Wolf, ed., *Progress in Optics*, Vol. VII (North-Holland, Amsterdam, '69). 141–168

J. P. Gordon, C. H. Wang, C. K. N. Patel, R. E. Slusher, + W. J. Tomlinson, Phys. Rev. 179, 294 ('69).

Circularly polarized plane wave: $E_+(z,t) = \mathcal{E}(z,t)e^{i[\omega t - kz - \phi(z)]}$

Wave equation: $\dfrac{\partial^2 E_+(z,t)}{\partial z^2} = \dfrac{\eta^2}{c^2}\dfrac{\partial^2 E_+(z,t)}{\partial t^2} + \dfrac{4\pi}{c^2}\dfrac{\partial^2 P_+(z,t)}{\partial t^2}$

Polarization: $P_+(z,t) = \int_{-\infty}^{\infty} g(\Delta\omega)[u(\Delta\omega,z,t) - iv(\Delta\omega,z,t)]e^{i[\omega t - kz - \phi(z)]}\, d(\Delta\omega)$

$$\frac{\partial E_+}{\partial z} = \frac{\partial \mathcal{E}}{\partial z}e^{i-} + i\mathcal{E}\left(-k - \frac{\partial\phi}{\partial z}\right)e^{i-}$$

$$\frac{\partial^2 E_+}{\partial z^2} = \left[\frac{\partial^2\mathcal{E}}{\partial z^2} - 2i\left(k + \frac{\partial\phi}{\partial z}\right)\frac{\partial\mathcal{E}}{\partial z} + (-1)\mathcal{E}\left(k + \frac{\partial\phi}{\partial z}\right)^2\right]e^{i-}$$

$$\frac{\partial E_+}{\partial t} = \left(\frac{\partial\mathcal{E}}{\partial t} + i\omega\mathcal{E}\right)e^{i-}$$

$$\frac{\partial^2 E_+}{\partial t^2} = \left(\frac{\partial^2\mathcal{E}}{\partial t^2} + 2i\omega\frac{\partial\mathcal{E}}{\partial t} - \omega^2\mathcal{E}\right)e^{i-}$$

$$\frac{\partial^2 P_+}{\partial t^2} = \int g\left[\underbrace{\frac{\partial^2}{\partial t^2}(u - iv) + 2i\omega\frac{\partial}{\partial t}(u - iv)}_{\ll} - \omega^2 (u - iv)\right]e^{i-}\, d(\Delta\omega)$$

SVEA

a ——

b ——

$$\cancel{\frac{\partial^2\mathcal{E}}{\partial z^2}} - 2i\left(k + \cancel{\frac{\partial\phi}{\partial z}}\right)\frac{\partial\mathcal{E}}{\partial z} - \mathcal{E}\left[\overset{(1)}{\cancel{k^2}} + 2k\frac{\partial\phi}{\partial z} + \cancel{\left(\frac{\partial\phi}{\partial z}\right)^2}\right] =$$

SVEA

$$\frac{\eta^2}{c^2}\left(\cancel{\frac{\partial^2\mathcal{E}}{\partial t^2}} + 2i\omega\frac{\partial\mathcal{E}}{\partial t} - \overset{(1)}{\cancel{\omega^2}}\mathcal{E}\right) + \frac{4\pi}{c^2}(-\omega^2)\int g(u - iv)\, d(\Delta\omega)$$

$$\boxed{\begin{aligned}
\frac{\partial\mathcal{E}}{\partial z} &= -\frac{\eta}{c}\frac{\partial\mathcal{E}}{\partial t} - \frac{2\pi\omega}{\eta c}\int_{-\infty}^{\infty} v(\Delta\omega,z,t)\,g(\Delta\omega)\,d(\Delta\omega)\\
\frac{\partial\phi(z)}{\partial z}\mathcal{E} &= \frac{2\pi\omega}{\eta c}\int_{-\infty}^{\infty} u(\Delta\omega,z,t)\,g(\Delta\omega)\,d(\Delta\omega)\\
\dot u &= v\Delta\omega - u/T_2' \qquad\qquad \Delta\omega = \omega_0 - \omega_{laser}\\
\dot v &= -u\Delta\omega - \kappa\mathcal{E}w - v/T_2'\\
\dot w &= \kappa\mathcal{E}v - (w - w_0)/T_1
\end{aligned}}$$

COUPLED MAXWELL-BLOCH EQUATIONS

$w = N_p\langle\sigma_z\rangle = N_p(\rho_{aa} - \rho_{bb})$

$u = \frac{N_p}{2}\langle\sigma_x\rangle = \frac{N_p}{2}(\rho_{ab} + \rho_{ba})$

$v = -\frac{N_p}{2}\langle\sigma_y\rangle = \frac{N_p}{2}i(\rho_{ba} - \rho_{ab})$

$u - iv = N_p\rho_{ba}$

$$\sigma_x = \begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix} \qquad \sigma_y = \begin{pmatrix}0 & -i\\ +i & 0\end{pmatrix} \qquad \sigma_z = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}$$

$$w = N_p\langle\sigma_z\rangle = N_p\begin{pmatrix}a^* & b^*\end{pmatrix}\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}\begin{pmatrix}a\\ b\end{pmatrix} = N_p\begin{pmatrix}a^* & b^*\end{pmatrix}\begin{pmatrix}a\\ -b\end{pmatrix}$$
$$= N_p(a^*a - b^*b)$$

$$\rho = |\psi\rangle\langle\psi| = \begin{pmatrix}a\\ b\end{pmatrix}\begin{pmatrix}a^* & b^*\end{pmatrix} = \begin{pmatrix}aa^* & ab^*\\ ba^* & bb^*\end{pmatrix} = \begin{pmatrix}\rho_{aa} & \rho_{ab}\\ \rho_{ba} & \rho_{bb}\end{pmatrix}$$

$$w = N_p(\rho_{aa} - \rho_{bb})$$

$$w = N_p\langle\sigma_z\rangle = N_p\,\mathrm{Tr}(\rho\sigma_z) = N_p\,\mathrm{Tr}\begin{pmatrix}\rho_{aa} & \rho_{ab}\\ \rho_{ba} & \rho_{bb}\end{pmatrix}\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}$$
$$= N_p\,\mathrm{Tr}\begin{pmatrix}\rho_{aa} & -\rho_{ab}\\ \rho_{ba} & -\rho_{bb}\end{pmatrix} = N_p(\rho_{aa} - \rho_{bb})$$

$$u = \frac{N_p}{2}\langle\sigma_x\rangle = \frac{N_p}{2}\mathrm{Tr}\begin{pmatrix}\rho_{aa} & \rho_{ab}\\ \rho_{ba} & \rho_{bb}\end{pmatrix}\begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix} = \frac{N_p}{2}\mathrm{Tr}\begin{pmatrix}\rho_{ab} & \rho_{aa}\\ \rho_{bb} & \rho_{ba}\end{pmatrix}$$
$$= \frac{N_p}{2}(\rho_{ab} + \rho_{ba})$$

$$v = -\frac{N_p}{2}\langle\sigma_y\rangle = \frac{N_p}{2}\mathrm{Tr}\begin{pmatrix}\rho_{aa} & \rho_{ab}\\ \rho_{ba} & \rho_{bb}\end{pmatrix}\begin{pmatrix}0 & i\\ -i & 0\end{pmatrix} = \mathrm{Tr}\frac{N_p}{2}\begin{pmatrix}-i\rho_{ab} & i\rho_{aa}\\ -i\rho_{bb} & i\rho_{ba}\end{pmatrix}$$
$$= \frac{N_p}{2}i(\rho_{ba} - \rho_{ab})$$

For an arbitrary $\mathcal{E}(0,t)$, the coupled Maxwell-Bloch equations can be solved numerically and $\mathcal{E}(z,t)$ determined. For no-loss cases $(T_1 = T_2' = \infty)$ the $\mathcal{E}(z,t)$ settle down in a length $z \simeq 5\alpha^{-1}$ to steady-state pulse shapes with areas $A = \kappa\int_{-\infty}^{\infty}\mathcal{E}(z,t)\,dt \simeq 2\pi, 4\pi, 6\pi, \ldots$ in agreement with the area theorem.

Except for the N_p normalization, these definitions differ from Brewer's only in that v here is the negative of Brewer's.

AREA THEOREM

If $\Delta\omega = 0$ + $T_1 = T_2' = \infty$, then

$$u = 0$$
$$\dot{v} = -\kappa\mathcal{E}w$$
$$\dot{w} = \kappa\mathcal{E}v$$

$$\ddot{v} = -\kappa^2\mathcal{E}^2 v \quad + \quad \ddot{w} = \kappa\mathcal{E}\dot{v} = -\kappa^2\mathcal{E}^2 w$$

$$w = A\cos\kappa\mathcal{E}t + B\sin\kappa\mathcal{E}t \qquad \text{if } \mathcal{E} = \text{const}$$

$$w(0) = -N_p = A$$
$$\dot{w} = -\kappa\mathcal{E}A\sin\kappa\mathcal{E}t + \kappa\mathcal{E}B\cos\kappa\mathcal{E}t$$
$$\dot{w}(0) = \kappa\mathcal{E}v(0) = 0 \Rightarrow B = 0$$

$$\frac{w}{N_p} = -\cos\kappa\mathcal{E}t \; ; \quad \frac{v}{N_p} = \sin\kappa\mathcal{E}t$$

but if $\mathcal{E} = \mathcal{E}(t)$, then

$$\frac{w}{N_p} = -\cos\varphi(z,t)$$
$$\varphi(z,t) \equiv \kappa\int_{-\infty}^{t}\mathcal{E}(z,t')\,dt'$$

$$\dot{w} = N_p(\sin\varphi)\,\kappa\mathcal{E} = \kappa\mathcal{E}v$$

$$v = (\sin\varphi)N_p$$

$$\dot{v} = N_p(\cos\varphi)\,\kappa\mathcal{E} = -\kappa\mathcal{E}w \quad ✓$$

$$\kappa\int_{-\infty}^{\infty}\left[\frac{\partial\mathcal{E}}{\partial z} = -\frac{\eta}{c}\frac{\partial\mathcal{E}}{\partial t} - \frac{2\pi\omega}{\eta c}\int_{-\infty}^{\infty} v(\Delta\omega, z, t')\,g(\Delta\omega)\,d(\Delta\omega)\right]dt'$$

$t \equiv T \to \infty$ so $\mathcal{E}(z,T) = 0$

$$\Rightarrow \quad \frac{d\theta(z)}{dz} = -\frac{2\pi\kappa\omega}{\eta c}\int_{-\infty}^{\infty}\int_{-\infty} g(\Delta\omega)\,v(\Delta\omega, z, t')\,d(\Delta\omega)\,dt'$$

$$\theta(z) \equiv \kappa\int_{-\infty}^{\infty}\mathcal{E}(z,t')\,dt'$$

$\mathcal{E} \to 0$ does not necessitate each $u(\Delta\omega, z, t)$ + $v(\Delta\omega, z, t)$ vanishing, but only that they destructively interfere because of the range of spectral frequencies $\Delta\omega$.

S. L. McCall & E. L. Hahn Phys Rev 183, 457 ...

In general

$$u(\Delta\omega, z, t) = u(\Delta\omega, z, T_0)\cos(\Delta\omega t') + v(\Delta\omega, z, T_0)\sin\Delta\omega t'$$

where $t = T_0 + t'$ with T_0 an arbitrary time origin having properties of T above, with $t' > 0$ measured w.r. to it

$$v = \dot{u}/\Delta\omega$$

$$\int v\,dt \sim u/\Delta\omega$$

$$\therefore \frac{d\theta(z)}{dz} = -\frac{2\pi\kappa\omega}{\eta c}\int_{-\infty}^{\infty}\left[\frac{g(\Delta\omega)}{\Delta\omega}\right]\left[u(\Delta\omega, z, T_0)\cos\Delta\omega t' + v(\Delta\omega, z, T_0)\sin\Delta\omega t'\right]d(\Delta\omega)$$

$$\int\frac{g(\Delta\omega)}{\Delta\omega}u(\Delta\omega, z, T_0)\cos\Delta\omega t' \simeq g(0)u(0,z,T_0)\underbrace{\int_{-\infty}^{\infty}\frac{\cos(\Delta\omega t')}{\Delta\omega}d(\Delta\omega)}_{\text{complex integrat.} = 0}$$

or argue $g(-\Delta\omega) = g(\Delta\omega)$ and $u(-\Delta\omega) = -u(\Delta\omega)$, $\therefore \int = \lim_{t' \to \infty} \dfrac{g(0)a_1\sin\Delta\omega t'}{t'}\Big|_{\Delta\omega}$

$u \propto a_1\Delta\omega + a_2(\Delta\omega)^3$

$$\int g(\Delta\omega)\,v(\Delta\omega, z, T_0)\underbrace{\frac{\sin\Delta\omega t'}{\Delta\omega}}_{= \delta(\Delta\omega)}d(\Delta\omega) = \pi\,g(0)\,v(0,z,T_0)$$

$$v(0,z,T_0) = N_p\sin\varphi(z,T_0) = N_p\sin\theta(z)$$

$$\therefore \boxed{\frac{d\theta(z)}{dz} = -\frac{\alpha}{2}\sin\theta(z)} \qquad \text{FAMOUS AREA THEOREM}$$

$$\alpha = \frac{8\pi^2 N_p p^2\omega g(0)}{\eta\hbar c}$$

weak light pulses $\Rightarrow \theta(z) \ll 1 \Rightarrow \dfrac{d\theta}{dz} \simeq -\dfrac{\alpha}{2}\theta \Rightarrow \ln\theta = -\dfrac{\alpha}{2}z$

$$\theta = \theta_0 e^{-\alpha z/2}$$

$$\theta = \kappa\int\mathcal{E}\,dt$$

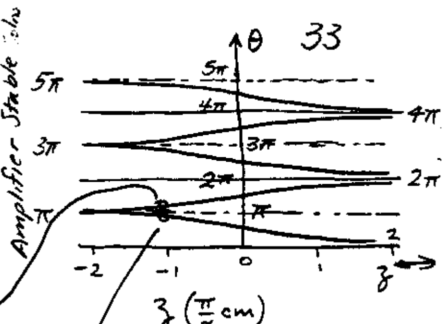
$$\mathcal{E} \sim \theta \Rightarrow \mathcal{E}(z) = \mathcal{E}(0)e^{-\alpha z/2}$$

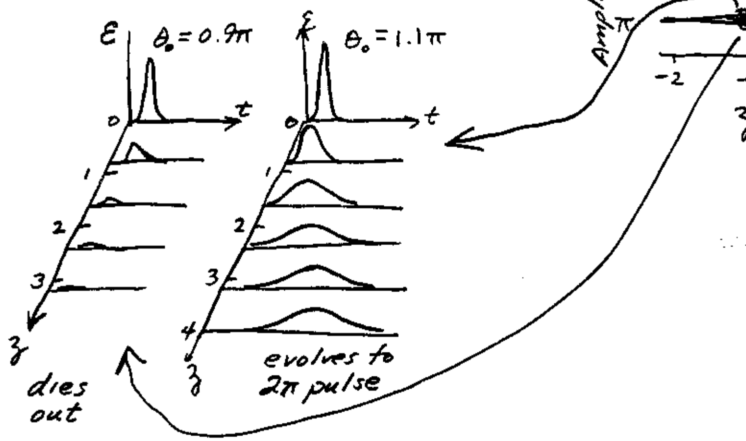
... Law for absorption

... the ... regime

General solution of area thm.

$$\theta(z) = 2\tan^{-1}\left[\tan\frac{\theta_0}{2}\, e^{-\alpha z/2}\right]$$





$$\frac{d\theta(z)}{dz} = -\frac{\alpha}{2}\sin\theta(z) \longrightarrow 0$$

$$\theta(z) = 0, 2\pi, 4\pi, \ldots$$

Solitons: pulses propagate with no further change in shape or pulse area.

Of great interest to mathematicians; applicable to many kinds of waves — water, acoustic, optical, etc.

See R.K. Bullough, "Solitons", in Interaction of Radiation with Condensed Matter, Vol. I, Intl. Atomic Energy Agency, Vienna, 1977.

Hyperbolic-Secant Solution for $\mathcal{E}(z,t)$

Sharp-line: $$\underbrace{\frac{\partial \mathcal{E}}{\partial z} = -\frac{1}{V}\frac{\partial \mathcal{E}}{\partial t}}_{\text{traveling wave}} = -\frac{2\pi\omega}{\eta c}\int_{-\infty}^{\infty} g(\Delta\omega)\, v(\Delta\omega, z, t)\, d(\Delta\omega) - \frac{\eta}{c}\frac{\partial \mathcal{E}}{\partial t}$$

$$g(\Delta\omega) = \delta(\Delta\omega) \quad + \quad v(\omega, z, t) = N p \sin\varphi$$ p. 97

$$\Rightarrow \frac{\partial \mathcal{E}}{\partial t} = \frac{2\pi\omega N p}{\eta c}\sin\varphi \Big/ \left(\frac{1}{V} - \eta/c\right)$$

$$\frac{\partial \varphi}{\partial t} = \kappa \mathcal{E} \qquad \text{from } \varphi \equiv \kappa\int_{-\infty}^{t} \mathcal{E}(t')\,dt'$$

$$\frac{\partial \mathcal{E}}{\partial t} = \frac{1}{\kappa}\frac{\partial^2 \varphi}{\partial t^2} = \frac{2\pi\omega N p}{\eta c\left(\frac{1}{V} - \eta/c\right)}\sin\varphi$$

$$\frac{\partial^2 \varphi}{\partial t^2} = \frac{1}{\tau^2}\sin\varphi \qquad \frac{1}{V} = \eta/c + \frac{2\pi\kappa\omega N p \tau^2}{\eta c}$$

$$\frac{\partial \mathcal{E}}{\partial t} = \frac{\partial \mathcal{E}}{\partial \varphi}\frac{\partial \varphi}{\partial t} = \kappa \mathcal{E}\frac{\partial \mathcal{E}}{\partial \varphi} = A\sin\varphi \qquad A = \frac{2\pi\omega N p}{\eta c\left(\frac{1}{V} - \eta/c\right)} = \frac{1}{\kappa\tau^2}$$

$$\kappa \mathcal{E}\, d\mathcal{E} = -A\, d(\cos\varphi)$$

$$\kappa \mathcal{E}^2/2 = -A(\cos\varphi - 1) = 2A\sin^2\varphi/2$$

$$\mathcal{E} = 2\sqrt{\frac{A}{\kappa}}\sin\varphi/2 = \underline{\underline{\frac{2}{\kappa\tau}\sin\varphi/2}}$$

$$q \equiv \kappa\mathcal{E}\tau/2;$$

$$\frac{\partial \mathcal{E}}{\partial t} = \frac{2}{\kappa\tau}(\cos\varphi/2)\frac{\partial \varphi/2}{\partial t} = \frac{\kappa\mathcal{E}}{\kappa\tau}\cos\varphi/2 = \frac{\mathcal{E}}{\tau}\sqrt{1-\sin^2\varphi/2} = \frac{\mathcal{E}}{\tau}\sqrt{1-\frac{\kappa^2\mathcal{E}^2\tau^2}{4}}$$

$$\dot{q} = \frac{q}{\tau}\sqrt{1-q^2}$$

$$\frac{dt}{\tau} = \frac{dq}{q\sqrt{1-q^2}} = -d(\operatorname{sech}^{-1} q) \qquad \text{Math tables}$$

$$\operatorname{sech}(-t/\tau) = q = \kappa\mathcal{E}\tau/2 \qquad \operatorname{sech} x = \frac{2}{e^x + e^{-x}} = \operatorname{sech}(-x)$$

$$\boxed{\mathcal{E}(z,t) = \frac{2}{\kappa\tau}\operatorname{sech}\left(\frac{t}{\tau} - \frac{z}{\tau V}\right)}$$ "2π" pulse:

$$\text{since } \frac{\partial \mathcal{E}}{\partial z} = -\frac{1}{V}\frac{\partial \mathcal{E}}{\partial t} \qquad \kappa\int_{-\infty}^{\infty}\mathcal{E}(z,t')\,dt' = 2\pi$$

Equivalent to $\varphi = 4\tan^{-1} e^{\frac{1}{\tau}(t - z/V)}$

Inhom. broadening

$$v(\Delta\omega, z, t) = v(0, z, t)\, f(\Delta\omega)$$

$$f(-\Delta\omega) = f(\Delta\omega), \quad f(0) = 1$$

4π analytic solution; mathematic[al] discussions, ...

George Lamb, Rev. Mod. Phys. 43, 99 (1971).

Assume that v factorizes:

$$v(\Delta\omega, z, t) = f(\Delta\omega)\, v(0, z, t) = f N p \sin\varphi$$

implying that off-resonance dipoles respond to $\mathcal{E}$ in the same way as the resonant dipoles, but perhaps with a detuning-dependent reduction in amplitude.

$$\dot w = \kappa \mathcal{E} v = \dot\varphi v = f N p \dot\varphi \sin\varphi = -f N p \frac{d}{dt}(\cos\varphi)$$

$$\therefore\ w = -fNp(\cos\varphi - 1) - Np$$

$$\dot u = (\Delta\omega) v$$

$$\dot v = -u\Delta\omega - \kappa\mathcal{E} w$$

$$\Delta\omega\, \dot u = (\Delta\omega)^2 v = fNp(\Delta\omega)^2 \sin\varphi$$

$$= -\ddot v - \kappa\mathcal{E}\dot w - \kappa\dot{\mathcal{E}} w$$

$$= -\ddot v - \dot\varphi \dot w - \ddot\varphi w$$

$$\dot v = fNp\,\dot\varphi\cos\varphi$$

$$\ddot v = fNp(\ddot\varphi\cos\varphi - \dot\varphi^2\sin\varphi)$$

$$\dot w = + fNp(\sin\varphi)\dot\varphi$$

$$\cancel{fNp}(\Delta\omega)^2\sin\varphi = \cancel{fNp}\left[\cancel{\dot\varphi^2\sin\varphi} - \cancel{\ddot\varphi\cos\varphi} - \cancel{\dot\varphi^2\sin\varphi} + \ddot\varphi(\cancel{\cos\varphi} - 1) + \ddot\varphi/f\right]$$

$$(\Delta\omega)^2 = \ddot\varphi\left(\tfrac{1}{f} - 1\right)/\sin\varphi$$

$$\ddot\varphi = \frac{(\Delta\omega)^2 f}{1-f}\sin\varphi$$

Since neither $\ddot\varphi$ nor $\sin\varphi$ depends upon $\Delta\omega$, then neither can

$$\frac{(\Delta\omega)^2 f(\Delta\omega)}{1 - f(\Delta\omega)} \equiv \frac{1}{\tau^2} \qquad \therefore\ f(\Delta\omega) = \frac{1}{1 + (\Delta\omega)^2\tau^2}$$

Energy loss:

$$\Delta W = \frac{N\hbar\omega/2}{Np}\Delta w = \frac{N\hbar\omega}{2} + \int_{-\infty}^{\infty} \frac{\hbar\omega}{2p} w(\Delta\omega, z, t=\infty)\, g(\Delta\omega)\, d(\Delta\omega)$$

$$\xrightarrow[\text{"}2\pi\text{" sech}]{} 0$$

$\Rightarrow$ u, v, w start out from $(0,0,-1)$, evolve, and then return exactly to $(0,0,-1)$, <u>independent</u> of the off-resonance

parameter $\Delta\omega$:

$$u(\Delta\omega, z, t) = (-\dot v - \kappa\mathcal{E} w)/\Delta\omega$$

$$= \{-fNp\dot\varphi\cancel{\cos\varphi} + \dot\varphi[fNp(\cancel{\cos\varphi} - 1) + Np]\}/\Delta\omega$$

$$= fNp\left(\tfrac{1}{f} - 1\right)\dot\varphi/\Delta\omega = Np\kappa\mathcal{E}(1-f)/\Delta\omega = Np\kappa\left(\frac{2}{\tau\kappa}\sin\varphi/2\right)\frac{\Delta\omega\,\tau^2}{1 + (\Delta\omega\tau)^2}$$

$$u(\Delta\omega, z, t) = \frac{2Np\,\Delta\omega\,\tau\sin\varphi/2}{1 + (\Delta\omega\tau)^2} = \frac{Np(\Delta\omega)\tau^2\kappa\mathcal{E}}{1 + (\Delta\omega\tau)^2}$$

$$v(\Delta\omega, z, t) = \frac{Np\sin\varphi}{1 + (\Delta\omega\tau)^2} = \frac{Np\tau\kappa\mathcal{E}\cos\varphi/2}{1 + (\Delta\omega\tau)^2}$$

$$w(\Delta\omega, z, t) = Np\left[\frac{2\sin^2\varphi/2}{1 + (\Delta\omega\tau)^2} - 1\right]$$

Fig. 13-2 in BSL

$$\sin\frac{\varphi}{2} = \operatorname{sech}\left[\frac{1}{\tau}(1 - z/V)\right] = \frac{\tau}{2}\kappa\mathcal{E}$$

So, <u>independent of $\Delta\omega$</u>, each dipole makes one & only one revolution during a 2π pulse. Note that this is in striking contrast to the rotation angle $\theta(\Delta\omega, z, t) = \int_{-\infty}^{t}[\kappa^2\mathcal{E}^2 + (\Delta\omega)^2]^{1/2}$ for a constant amplitude square pulse. The sech has the very special shape required for all dipoles to precess together — as is essential if there is to be no energy loss to the absorbers. Hence the name <u>self-induced transparency</u> — a pulse through resonant exchange of energy with the absorbers is able to propagate without loss of energy or change of shape through a medium highly absorbing for low area pulses.

<u>Pulse velocity</u>:

$$\frac{\partial\mathcal{E}}{\partial z} = -\frac{1}{V}\frac{\partial\mathcal{E}}{\partial t} = -\frac{2\pi\omega}{\eta c}\int g v\, d(\Delta\omega) - \frac{\eta}{c}\frac{\partial\mathcal{E}}{\partial t}$$

$$\frac{1}{V} = \frac{\eta}{c} + \frac{2\pi\omega}{\eta c}\int g v\, d(\Delta\omega)\Big/\frac{\partial\mathcal{E}}{\partial t} = \eta/c + \frac{2\pi\omega Np\sin\varphi}{\eta c\,\partial\mathcal{E}/\partial t}\int\frac{g}{1 + (\Delta\omega\tau)^2}\, d(\Delta\omega)$$

$$\mathcal{E} = \frac{2}{\kappa\tau}\sin\varphi/2 \qquad \frac{\partial\mathcal{E}}{\partial t} = \frac{2}{\kappa\tau}\frac{\dot\varphi}{2}\cos\frac{\varphi}{2} = \frac{\mathcal{E}}{\tau}\cos\frac{\varphi}{2} = \frac{2}{\kappa\tau^2}\sin\frac{\varphi}{2}\cos\frac{\varphi}{2} = \frac{\sin\varphi}{\kappa\tau^2}$$

$$\therefore\ \frac{2\pi\omega Np\sin\varphi}{\eta c\,\partial\mathcal{E}/\partial t} = \frac{2\pi\omega Np\kappa\tau^2}{\eta c} = \frac{\alpha}{2\pi g(0)}$$

$$\boxed{\frac{1}{V} = \frac{\eta}{c} + \frac{\alpha\tau^2}{2\pi g(0)}\int_{-\infty}^{\infty}\frac{g(\Delta\omega)\, d(\Delta\omega)}{1 + (\Delta\omega\tau)^2}}$$

$$T_2^* \ll \tau, \quad \alpha\tau \gg \eta/c \Rightarrow \frac{1}{V} \approx \frac{\alpha\tau}{2}$$

$$V = \frac{L}{\tau_d} \Rightarrow \boxed{\tau_d \approx \frac{\alpha L\tau}{2}}$$

Delay time — roughly one pulse width per α^{-1} length

Dispersion relation:

$$\mathcal{E}\frac{\partial\phi}{\partial z} = \frac{2\pi\omega}{\eta c}\int u\, g\, d(\Delta\omega)$$

$$u = \frac{N p \tau^2 \kappa \mathcal{E}}{(1+\Delta\omega\tau)^2}\,\Delta\omega$$

$$\frac{\partial\phi}{\partial z} = \frac{2\pi\omega}{\eta c} N p \kappa \int \frac{\Delta\omega\, g(\Delta\omega)}{(\Delta\omega)^2 + 1/\tau^2}\, d(\Delta\omega)$$

$$\alpha = \frac{8\pi^2 N p^2 \omega g(0)}{\eta \hbar c}$$

$$\boxed{\frac{\partial\phi}{\partial z} = \frac{\alpha}{2\pi g(0)}\int \frac{\Delta\omega}{(\Delta\omega)^2 + 1/\tau^2}\, g(\Delta\omega)\, d(\Delta\omega)}$$

$$E(z,t) = \mathcal{E}(z,t)\, e^{i(\omega t - kz - \phi)}$$

$$k' = k + \frac{\partial\phi}{\partial z}$$

$$k' - k = \frac{\partial\phi}{\partial z}$$

Neither the velocity formula nor the dispersion relation involve $\hbar$ directly. Only in $\mathcal{E} = \frac{2}{\kappa\tau}\,\mathrm{sech}\,\frac{1}{\tau}(1 - z/V)$

$$= \frac{\hbar}{p\tau}\,\mathrm{sech}\,\frac{1}{\tau}(1 - z/V)$$

is the evidence of the nonclassical + nonlinear foundation of the effect seen.

Propagation of small area pulses under assumption of weak excitation, i.e., linear response is treated on p. 104. For detailed discussion see

M. D. Crisp, Phys. Rev. A $\underline{1}$, 1604 (1970).
Opt. Commun. $\underline{4}$, 199 (1971).
Appl. Optics $\underline{11}$, 1124 (1972).

$$\dot{u} = v\Delta\omega - u/T_2' \; ; \; \dot{v} = -u\Delta\omega - v/T_2' - \kappa\mathcal{E}w \qquad \mathcal{E} \text{ real}$$

Linear approximation $\Rightarrow$ system excitation is always small
$\Rightarrow w \approx -Np$

$$\dot{u} - i\dot{v} \approx (u - iv)(i\Delta\omega - 1/T_2') - i\kappa E N p \qquad E \text{ complex}$$

$$\left(\frac{\partial}{\partial z} + \frac{\eta}{c}\frac{\partial}{\partial t}\right) E = -\frac{i2\pi\omega}{\eta c}\int (u - iv)\, g\, d(\Delta\omega)$$

Fourier decompose $E(z,t)$ and $P(t,z,\Delta\omega) \equiv u(t,z,\Delta\omega) - iv(t,z,\Delta\omega)$

$$E(z,t) = e(\nu,z)\, e^{i\nu t} \; ; \; P(t,z,\Delta\omega) = P(\nu,z,\Delta\omega)\, e^{i\nu t}$$

(assumes dipoles oscillate at driving frequency).

$$\left(\frac{\partial}{\partial z} + \frac{\eta}{c} i\nu\right) e(\nu,z)\, e^{i\nu t} = -\frac{i2\pi\omega}{\eta c} e^{i\nu t}\int P(\nu,z,\Delta\omega)\, g\, d(\Delta\omega)$$

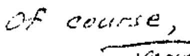
$$i\nu P(\nu,z,\Delta\omega)\cancel{e^{i\nu t}} \approx P(\nu,z,\Delta\omega)\cancel{e^{i\nu t}}(i\Delta\omega - 1/T_2') - i\kappa N p\, e(\nu,z)\cancel{e^{i\nu t}}$$

$$P(\nu,z,\Delta\omega) = \frac{\kappa e(\nu,z) N p}{\Delta\omega - \nu + i/T_2'}$$

$$\therefore \left[\frac{\partial}{\partial z} + \frac{\eta}{c} i\nu + \mathcal{P}(\nu)\right] e(\nu,z) = 0 \quad \text{where } \mathcal{P}(\nu) \equiv \frac{i8\pi^2 N p^2}{\eta\lambda\hbar}\int \frac{g(\Delta\omega)}{\Delta\omega - \nu + i/T_2'}\, d(\Delta\omega)$$

$$\therefore e(\nu,z) = e(\nu,0)\, e^{-i[\eta\nu/c - i\mathcal{P}(\nu)]z}$$

$$= e(\nu,0)\, e^{-i\eta\nu z/c}\, e^{-\mathcal{P}(\nu)z} = e(\omega,0)\, e^{-i\left[\frac{\eta\nu}{c} + \mathcal{P}_{Im}(\nu)\right]z}\, e^{-\mathcal{P}_{Re}(\nu)z}$$

$\Rightarrow$ Each Fourier component is reduced by the absorption coefficient, $\mathcal{P}_{Re}(\nu)$, at that frequency. So a pulse of arbitrary time and frequency profiles, can be analyzed in frequency space (Fourier-analyzed) and then multiplied by $e^{-\mathcal{P}_{Re}(\nu)z}$ at each frequency. This implies that a pulse with $\tau_p \ll \tau_{nat.}$ so that $\Delta\nu_p \gg \Delta\nu_{nat.}$ incident on a beam of atoms with $\Delta\nu_{abs.} = \Delta\nu_{nat.}$ will have a notch in frequency space removed: . Of course, the time shape must change if the frequency shape changes. Variation on

SR   SF

$$\dot u = v\,\Delta\omega - u/T_2'$$
$$\dot v = -u\,\Delta\omega - v/T_2' - \kappa\mathcal{E}w$$
$$\dot w = -(w-w_0)/T_1 + \kappa\mathcal{E}v$$

$$\frac{\partial\mathcal{E}}{\partial z} + \frac{1}{c}\frac{\partial\mathcal{E}}{\partial t} = -\frac{2\pi\omega}{c}\int g\,v\,d(\Delta\omega)$$

$$\Delta\omega = 0,\quad T_1 = T_2' = \infty \;\Rightarrow\; \dot u = 0,\; \dot v = -\kappa\mathcal{E}w,\; \dot w = \kappa\mathcal{E}v$$

$$\frac{\partial\mathcal{E}}{\partial z} + \frac{1}{c}\frac{\partial\mathcal{E}}{\partial t} = -\frac{2\pi\omega v}{c} \;\cdot\!= \frac{\partial\mathcal{E}}{\partial Z}$$

$$\theta = \frac{2p}{\hbar}\int_{-\infty}^{t}\mathcal{E}\,dt \;\Rightarrow\; \frac{\partial\theta}{\partial T} = 2p\mathcal{E}/\hbar$$

$$\frac{\partial^2\theta}{\partial Z\,\partial T} = \frac{2p}{\hbar}\frac{\partial\mathcal{E}}{\partial Z} = -\frac{4\pi\omega p}{\hbar c}\,v$$

$$\begin{cases} T = t - z/c;\; Z = z \\ \frac{\partial}{\partial t} = \frac{\partial}{\partial T} \\ \frac{\partial}{\partial z} = -\frac{1}{c}\frac{\partial}{\partial T} + \frac{\partial}{\partial Z}\end{cases}$$

$v = Np\sin\theta$, $W = -Np\cos\theta$ p. 97   initially in ground state

$$\frac{\partial^2\theta}{\partial Z\,\partial T} = -\frac{4\pi\omega n p_{m}^2\sin\theta}{\hbar c} = -\frac{\sin\theta}{\tau_R L}$$

$$\frac{1}{\tau_0} = \frac{4}{3}\frac{\omega^3 p_{MH}^2\,2}{\hbar c^3}$$ because $p_{Skribanowitz} = \sqrt{2}\,p_{McCall-Hahn}$

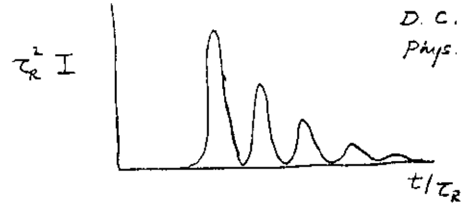
$$\therefore\; \frac{1}{\tau_R L} = \frac{4\pi\omega n p^2}{\hbar c} = \frac{4\pi\omega n}{\cancel{\hbar c}}\,\frac{3\cancel{\hbar c}\,c^2}{8\omega^3\tau_0} = \frac{3\pi\lambda^2 n}{8\pi^2\tau_0}$$

$$\therefore\; \boxed{\tau_R = \frac{8\pi}{3}\frac{\tau_0}{n\lambda^2 L}}$$

$$\boxed{\frac{\partial^2\theta}{\partial Z\,\partial T} = -\frac{\sin\theta}{\tau_R L}}$$ Initially in ground state; θ measured w.r.t. gd. state

$$\theta \to \varphi - \pi$$
$$\Rightarrow \boxed{\frac{\partial^2\varphi}{\partial Z\,\partial T} = \frac{\sin\varphi}{\tau_R L}}$$ sine-Gordon equation

The sine-Gordon equation (or, equivalently, the (corrected) coupled M-B eqs.) can be integrated numerically showing a ringing in the emission for the range of experiments performed or reasonably conceived:



D. C. Burnham & R. Y. Chiao, Phys. Rev. 188, 667 (1969).

Before N. Skribanowitz, I. P. Herman, J. C. MacGillivray, and M. S. Feld, Phys. Rev. Lett. 30, 309 (1973), propagation effects were discussed occasionally but always neglected in the final analysis. A single pulse sech² intensity was then obtained, as can be seen rather quickly:

$$\varphi \equiv \kappa\int_{-\infty}^{t}\mathcal{E}(t')\,dt'$$

$$\dot\varphi = \kappa\mathcal{E} \quad \text{but} \quad \frac{\partial\mathcal{E}}{\partial Z} = -\frac{2\pi\omega v}{c}$$

from p. 97 with $\Delta\omega = 0$, $T_1 = T_2' = \infty$, $v = -Np\sin\varphi$

No spatial variations $\Rightarrow v \neq v(z) \Rightarrow \mathcal{E}(L) = \frac{2\pi\omega Np}{c}L\sin\bar\varphi$; $\mathcal{E}(0) = 0$

$\bar\varphi$ av. over z $\bar{\mathcal{E}} \simeq \frac{\pi\omega NpL}{c}\sin\bar\varphi$

$$\dot{\bar\varphi} = \kappa\bar{\mathcal{E}} = \frac{\sin\bar\varphi}{2\tau_R}$$ or integrate sine-Gordon eq $\left.\frac{\partial\varphi}{\partial T}\right|_L - \left.\frac{\partial\varphi}{\partial T}\right|_0^{=0} \simeq \left(\frac{\sin\bar\varphi}{\tau_R L}\right)\frac{L}{2}$

$$\ddot{\bar\varphi} = \frac{\dot{\bar\varphi}\cos\bar\varphi}{2\tau_R} = \frac{\sin\bar\varphi\cos\bar\varphi}{(2\tau_R)^2} = \frac{\sin 2\bar\varphi}{2(2\tau_R)^2}$$

$$\ddot{\bar\varphi}_1 = \frac{\sin\bar\varphi_1}{(2\tau_R)^2},\quad \bar\varphi_1 = 2\bar\varphi \qquad \dot{\bar\varphi}_1 = 2\dot{\bar\varphi} = 2\kappa\bar{\mathcal{E}}$$

p. 100, if $\dot\varphi = \kappa\mathcal{E}$ & $\ddot\varphi = \frac{1}{\tau^2}\sin\varphi$ then $\mathcal{E} = \frac{2}{\kappa\tau}\sin\varphi/2 = \frac{2}{\kappa\tau}\,\mathrm{sech}\,t/\tau$

$$\therefore\; \bar{\mathcal{E}} = \frac{2}{(2\kappa)(2\tau_R)}\,\mathrm{sech}\!\left(\frac{t-\tau_0}{2\tau_R}\right)$$

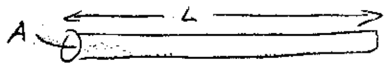
$$t = 0,\; \bar{\mathcal{E}} = \frac{1}{2\kappa\tau_R}\sin\bar\varphi_1(0)/2 = \frac{1}{2\kappa\tau_R}\underbrace{\sin\bar\varphi(0)}_{\approx\bar\varphi(0)\text{ for SF}} = \frac{1}{2\kappa\tau_R}\left(\frac{2}{e^{-\tau_0/2\tau_R} + e^{\tau_0/2\tau_R}}\right) \approx \frac{2e^{-\tau_0/2\tau_R}}{2\kappa\tau_R}$$

$\tau_0/\tau_R \approx 20$ to 100 usually

$$\therefore\; \boxed{\tau_0 \approx 2\tau_R\ln\frac{2}{\bar\varphi(0)}}$$

$$\boxed{I = \frac{c}{4\pi}\mathcal{E}^2 = \frac{N\hbar\omega}{\ldots}\,\mathrm{sech}^2\!\left(\frac{t-\tau_0}{2\tau_R}\right)} \sim N^2$$

Rough estimate of τ_R: one can show that the polarization excited by the first photon which travels down the pencil is large enough to dominate the evolution from that time onward in essentially a classical manner.



Assume an inverted pencil of Fresnel number $\mathcal{N} = \frac{A}{\lambda L}$ of about unity. If $\mathcal{N} > 1 \Rightarrow$ more than one transverse mode. If $\mathcal{N} < 1 \Rightarrow$ diffraction reduces lengthwise communication. The time for the first photon along the pencil is then roughly

$$\tau_R \approx \underbrace{\frac{\tau_0}{N}}_{N = \text{no. of atoms}} \underbrace{\left(\frac{4\pi L^2}{A}\right)}_{\text{probability of spontaneously emitted photon being in proper solid angle}}$$

$$\propto \frac{\tau_0}{nAL} \frac{4\pi L^2}{A} \xrightarrow[\mathcal{N} = \frac{A}{\lambda L} = 1]{} \frac{4\pi\tau_0 \cancel{L^2}}{nL\lambda^2 \cancel{L^2}} = \frac{4\pi\tau_0}{nL\lambda^2}$$

More careful calculations lead to $\boxed{\tau_R = \frac{8\pi\tau_0}{3n\lambda^2 L}}$ Superfluorescence Time

Cooperation time $\equiv$ value of τ_R for a sample of length such that light travels a sample length in a cooperation time

$$\Rightarrow \quad \tau_c = \frac{8\pi\tau_0}{3n\lambda^2(\tau_c c)} \quad \Rightarrow \quad \boxed{\tau_c^2 = \frac{8\pi\tau_0}{3n\lambda^2 L}\frac{L}{c} = \tau_R \tau_E}$$

where the escape time τ_E is simply L/c: $\boxed{\tau_E = L/c}$

So for near-ideal SF one wants:

	Escape		Cooperation		SF		Delay		Longitudinal, Transverse (Homog.), Dephasing (Inhom.)
①	τ_E	$<$	τ_c	$<$	τ_R	$<$	τ_D	$<$	T_1, T_2', T_2^*
	L/c		$\sqrt{\tau_E\tau_R}$		$\frac{8\pi\tau_0}{3n\lambda^2 L}$		$\approx \tau_R \ln N$		
	0.067	$<$	0.18	$<$	0.5	$<$	10	$<$	70, 80, 32 ns

for Cs 7P–7S, $\lambda = 2.931\,\mu m$, $L = 2$ cm, $\tau_0 = 551$ ns

② Excitation pulse τ_p $<<$ SF evolution time τ_D

$2 << 10$

} So details of excitation can be avoided in the analysis.

③ Fresnel number $\mathcal{N} = \frac{A}{\lambda L} \approx 1$ for single mode.

The beam area of the exciting pulse was adjusted so that $\mathcal{N} = 1$ for each L.

These conditions are discussed (with slight differences in definitions) by Bonifacio + Lugiato, Phys. Rev. A 11, 1507 (1975).