Optical Bloch Equations II

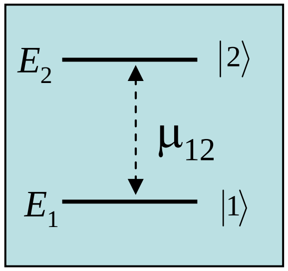
The Optical Bloch Equations (OBEs) describe the interaction of light and matter (atoms) including coherence

Equations of motion for elements of density matrix

Outline:

1) Write down OBEs
2) Rotating wave approximation (RWA)
3) Rabi Solution
4) Area theorem
5) Macroscopic Polarization
6) Bloch Vector
7) Perturbation Theory

Optical Bloch Equations (OBE)



Last time we developed the pieces needed to write the Optical Bloch Equations, which are the equations of motion for the elements of the density matrix

$$\dot{\rho} = -\frac{1}{2}[\Gamma\rho + \rho\Gamma] - \frac{i}{\hbar}[H,\rho]$$

where $\Gamma = \gamma_i \delta_{ij}$

write

$$H = H_0 + V$$

H_0 is the Hamiltonian for a 2-level system

$$H_0 = \begin{pmatrix} E_1 & 0 \\ 0 & E_2 \end{pmatrix}$$

V is the coupling to the EM field (later treat as small perturbation)

$$V = \underline{\boldsymbol{\mu}_{12}} E = E(t)\mu_{12}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$$

dipole moment operator

Plugging these into the equation of motion for ρ gives

$$\boxed{\begin{aligned}
\dot{\rho}_{11} &= -\gamma_1\rho_{11} + \tfrac{i}{\hbar}\mu_{12}E(t)(\rho_{12} - \rho_{21}) && (1)\\
\dot{\rho}_{22} &= -\gamma_2\rho_{22} - \tfrac{i}{\hbar}\mu_{12}E(t)(\rho_{12} - \rho_{21}) && (2)\\
\dot{\rho}_{12} &= -\gamma_{12}^T\rho_{12} + i\omega_0\rho_{12} - \tfrac{i}{\hbar}\mu_{12}E(t)(\rho_{22} - \rho_{11}) && (3)
\end{aligned}}$$

where

$$\omega_0 = \frac{E_2 - E_1}{\hbar}, \quad \gamma_{12}^T = \tfrac{1}{2}(\gamma_1 + \gamma_2) + \gamma_{ph}$$

Rotating Wave Approximation I

Write the electric field

$$E(t) = \tilde{E}(t)\cos(\omega t) = \tfrac{1}{2}\left(\hat{E}(t)e^{i\omega t} + \hat{E}^*(t)e^{-i\omega t}\right)$$

and

$$\rho_{12} = \hat{\rho}_{12}e^{i\omega t} \qquad \dot{\rho}_{12} = \dot{\hat{\rho}}_{12}e^{i\omega t} + i\omega\hat{\rho}_{12}e^{i\omega t}$$

equation (3) becomes

$$\dot{\hat{\rho}}_{12} = -\gamma_{12}^T\hat{\rho}_{12} + i(\omega_0 - \omega)\hat{\rho}_{12} - \tfrac{i}{2\hbar}\mu_{12}\left(\hat{E}(t) + \hat{E}^*(t)e^{-i2\omega t}\right)(\rho_{22} - \rho_{11})$$

assume at $t = 0$, $\rho_{22} = 0$, $\rho_{11} = N$ (i.e. all atoms in the ground state)

multiply by integrating factor $\exp\left[-\gamma_{12}^T t + i(\omega - \omega_0)t\right]$

$$\hat{\rho}_{12} = \frac{-iN}{2\hbar}\mu_{12}\int_0^t \left(\hat{E}(t')\exp\left[-\gamma_{12}^T t' + i(\omega - \omega_0)t'\right] + \hat{E}^*(t')\exp\left[-\gamma_{12}^T t' - i(\omega + \omega_0)t'\right]\right)dt'$$

if we assume $\hat{E}(t) = \hat{E}^*(t) = 1$ (CW field) we can do integral

Rotating Wave Approximation II

CW solution is

$$\rho_{12} = \frac{iN}{2\hbar}\mu_{12}\left(\frac{\exp\left[-\gamma_{12}^T t' + i\left(\omega-\omega_0\right)t'\right]}{-\gamma_{12}^T + i\left(\omega-\omega_0\right)} + \frac{\exp\left[-\gamma_{12}^T t' - i\left(\omega+\omega_0\right)t'\right]}{-\gamma_{12}^T - i\left(\omega+\omega_0\right)}\right)\Bigg|_{t'=0}^{t}$$

if $\omega \sim \omega_0$, ignore second term $\left(\omega+\omega_0 >> \omega-\omega_0\right)$

This is the rotating wave approximation: we only include one phasor of the electric field (physical picture for S → P transition)

Second term causes a shift of the apparent resonance (Bloch-Siegert shift), observable in infrared for broad resonances

Write OBEs in RWA (implicitly in rotating frame):

$$\dot{\rho}_{11} = -\gamma_1\rho_{11} + \tfrac{i}{2\hbar}\mu_{12}\left(\hat{E}^*(t)\rho_{12} - \hat{E}(t)\rho_{21}\right)$$
$$\dot{\rho}_{22} = -\gamma_2\rho_{22} - \tfrac{i}{2\hbar}\mu_{12}\left(\hat{E}^*(t)\rho_{12} - \hat{E}(t)\rho_{21}\right)$$
$$\dot{\rho}_{12} = -\gamma_{12}^T\rho_{12} + i(\omega_0 - \omega)\rho_{12} - \tfrac{i}{2\hbar}\mu_{12}\hat{E}(t)(\rho_{22} - \rho_{11})$$

However, care is needed that the RWA is not invoked too early

Rabi Solution I

- Ignore decay terms
- Assume $\hat{E}(t) = E$ (real and constant)
- Define the inversion $n = \rho_{22} - \rho_{11}$

Then

$$\dot{n} = -i\Omega_0(\rho_{12} - \rho_{21})$$

$$\dot{\rho}_{12} = i\Delta\omega\rho_{12} - \frac{i}{2}\Omega_0 n$$

where $\Delta\omega = \omega_0 - \omega$ and the Rabi frequency is $\Omega_0 = \frac{\mu_{12}E}{\hbar}$

split out real and imaginary parts of $\rho_{12} = a + ib$

$$\dot{n} = 2\Omega_0 b \qquad \dot{a} = -\Delta\omega b \qquad \dot{b} = \Delta\omega a - \frac{\Omega_0}{2}n$$

in matrix form

$$\begin{pmatrix} \dot{a} \\ \dot{b} \\ \dot{n} \end{pmatrix} = \begin{pmatrix} 0 & -\Delta\omega & 0 \\ \Delta\omega & 0 & -\frac{\Omega_0}{2} \\ 0 & 2\Omega_0 & 0 \end{pmatrix} \begin{pmatrix} a \\ b \\ n \end{pmatrix}$$

Rabi Solution II

Solution: $A = e^{-\lambda t}\begin{pmatrix} a_0 \\ b_0 \\ n_0 \end{pmatrix}$ where λ is an eigenvalue and $\begin{pmatrix} a_0 \\ b_0 \\ n_0 \end{pmatrix}$ the corresponding eigenvector

The characteristic equation is

$$-\lambda^3 - \lambda\Omega_0^2 - \Delta\omega^2\lambda = 0$$

solutions are $\lambda = 0, \quad \lambda = \pm i\sqrt{\Omega_0^2 + \Delta\omega^2} = \pm i\Omega(\Delta\omega)$

take $\Delta\omega = 0$ then $\Omega(\Delta\omega) \rightarrow \Omega_0$

$$\dot{n} = 2\Omega_0 b \qquad \dot{b} = -\frac{\Omega_0}{2} n$$

Initial conditions: n = -1 (system in ground state); $b = 0$ (no initial dipole)

Solutions:

$$n = -\cos\Omega_0 t$$ ⟵ Field causes population(inversion) to oscillate between ground and excited states as frequency Ω_0 (the Rabi frequency). This is called “Rabi flopping”

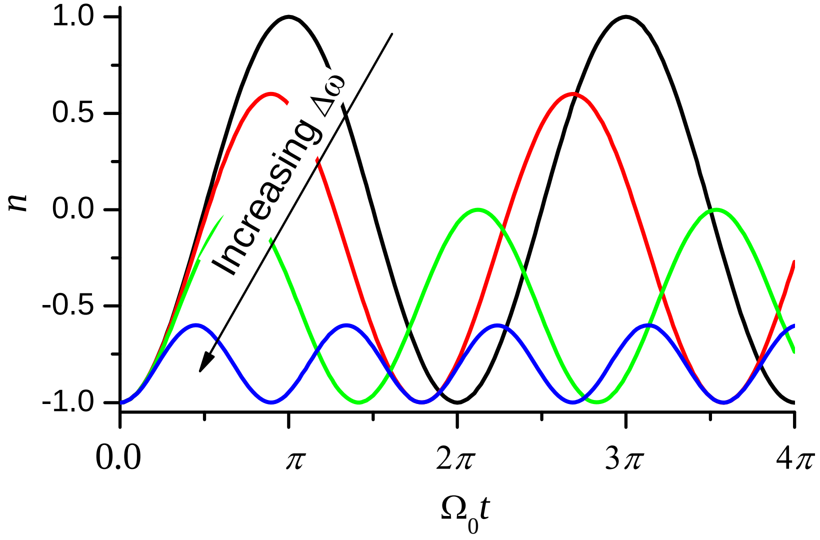
$$b = \frac{1}{2}\sin\Omega_0 t$$

Rabi Solution III

More general solution [assuming $a(0) = b(0) = 0$]

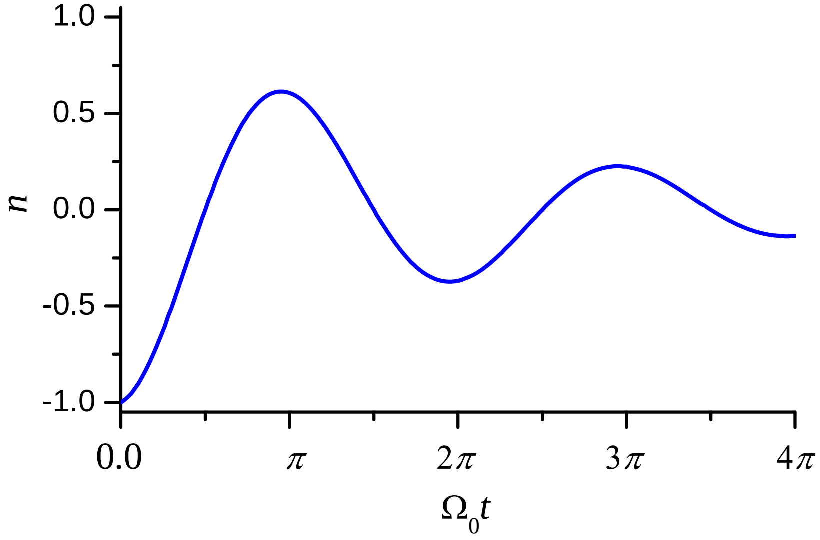
$$n(t) = n_0 \frac{\Delta\omega^2 + \Omega_0^2 \cos[\Omega(\Delta\omega)t]}{[\Omega(\Delta\omega)]^2}$$

for increasing $\Delta\omega$, oscillations in n get faster but smaller



A square resonant pulse with length $\Omega_0 t = \pi$ drives the system into the excited state, known as π-pulse

A square resonant pulse with length $\Omega_0 t = 2\pi$ drives the system into the excited state and then back to ground state, known as 2π-pulse



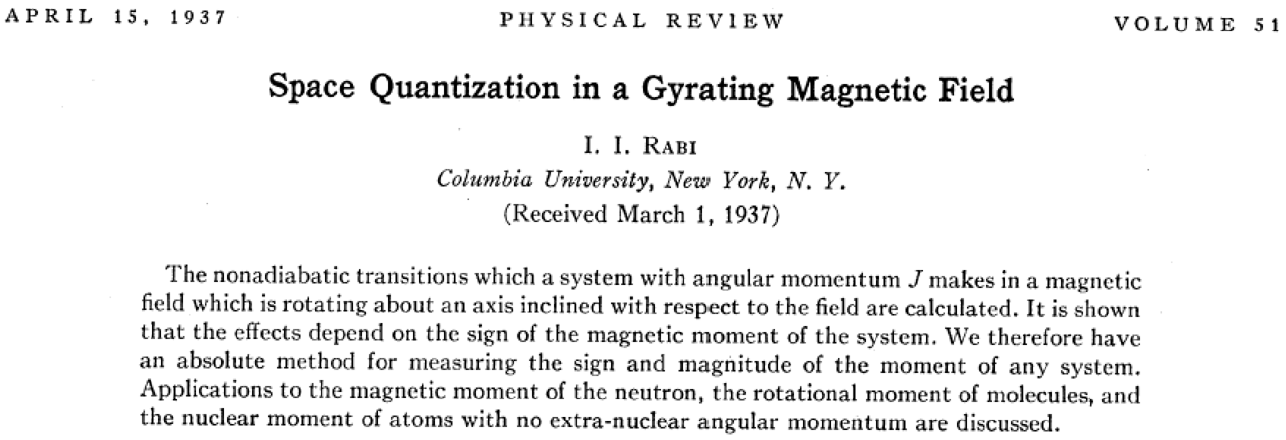
Dephasing causes the population to not return to zero

Generalized Rabi Frequency

$$\Omega(\Delta\omega) = \sqrt{\Omega_0^2 + \Delta\omega^2}$$

Rabi Flopping history

Rabi's original formulation was for magnetic resonance



APRIL 15, 1937 PHYSICAL REVIEW VOLUME 51

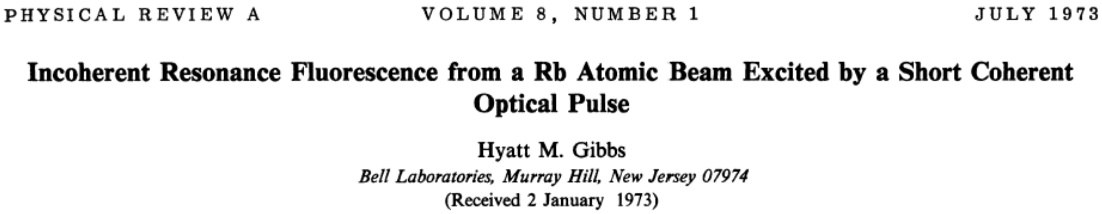
Space Quantization in a Gyrating Magnetic Field

I. I. RABI
Columbia University, New York, N. Y.

(Received March 1, 1937)

The nonadiabatic transitions which a system with angular momentum J makes in a magnetic field which is rotating about an axis inclined with respect to the field are calculated. It is shown that the effects depend on the sign of the magnetic moment of the system. We therefore have an absolute method for measuring the sign and magnitude of the moment of any system. Applications to the magnetic moment of the neutron, the rotational moment of molecules, and the nuclear moment of atoms with no extra-nuclear angular momentum are discussed.

Clear observation in optics by Gibbs

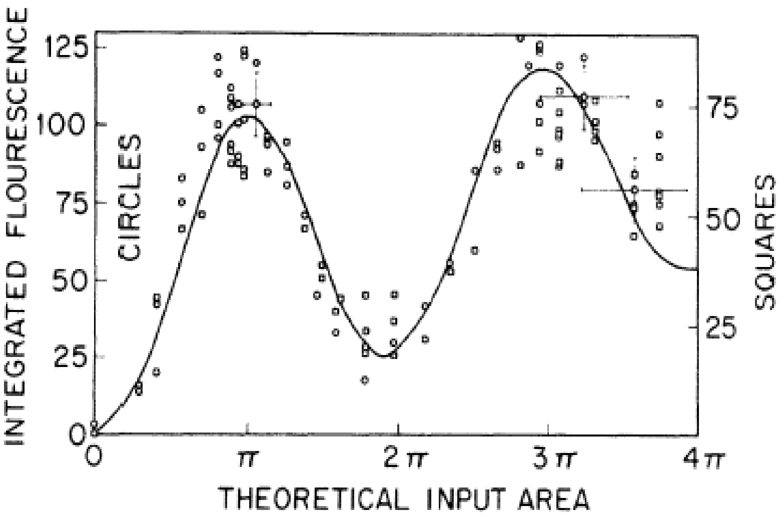


PHYSICAL REVIEW A VOLUME 8, NUMBER 1 JULY 1973

Incoherent Resonance Fluorescence from a Rb Atomic Beam Excited by a Short Coherent Optical Pulse

Hyatt M. Gibbs
Bell Laboratories, Murray Hill, New Jersey 07974

(Received 2 January 1973)

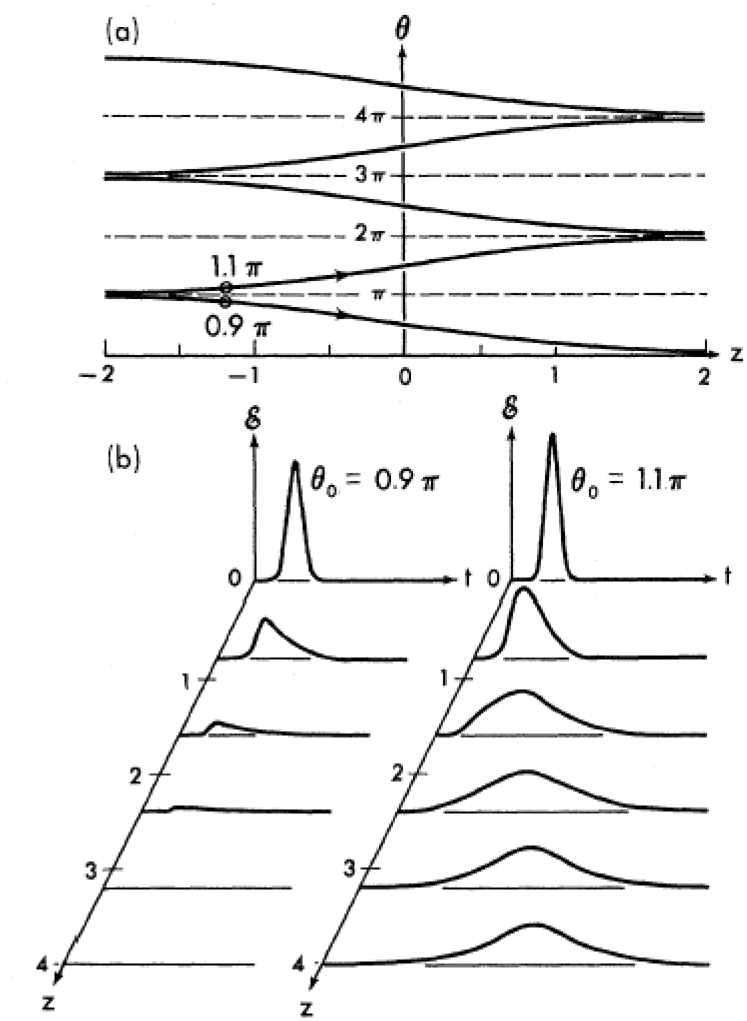


Area theorem

For pulses short compared to relaxation time, non-square pulses can be characterized by their area (z is position in medium)

$$A(t,z) = \mu_{12} \int_{-\infty}^{t} \hat{E}(t',z)\,dt'$$

Final state of OBEs matches that for a square pulse with same area



Propagation

Define total area

$$A_T(z) = \mu_{12} \int_{-\infty}^{\infty} \hat{E}(t',z)\,dt'$$

Classical

$$\frac{d}{dz} A_T(z) = -\frac{1}{2}|\alpha| A_T(z)$$ (Beer's law)

Quantum

$$\frac{d}{dz} A_T(z) = \frac{1}{2}\alpha \sin\left(A_T(z)\right)$$

Pulse with $A_T < \pi$ will decay, while $A_T < \pi$ evolves toward 2π

Pulse with area 2π is stable and evolves without loss: Self-induced transparency

Macroscopic Polarization

The polarization is $N\langle \boldsymbol{\mu}_{12} \rangle$

$$\begin{aligned} P = N\langle \boldsymbol{\mu}_{12} \rangle = N\,\mathrm{Tr}(\boldsymbol{\mu}_{12}\boldsymbol{\rho}) &= N\mu_{12}(\rho_{12} + \rho_{21}) \\ &= 2N\mu_{12}\,\mathrm{Re}(\rho_{12}) \end{aligned}$$

Identify off diagonal elements with a polarization

Recall that the polarization is the driving term in Maxwell's equations.

Bloch Vector

Because the Bloch equations were originally derived for NMR for a real spin, the OBE are often discussed in terms of a pseudo-spin or **Bloch vector** picture

usually in frame rotating with incident field

3 coordinates are (a, b, n)

π-pulses & Bloch vector

0 detuning π pulse:

(0,0,-1) → (1,0,0) → (0,0,1)

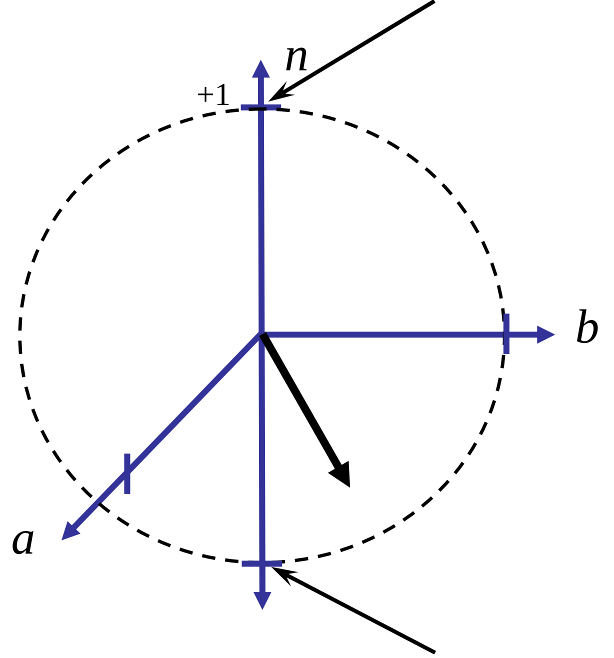
0 detuning 2π pulse

(0,0,-1) → (0,0,1) → (0,0,-1)

detuning tilts plane of motion

less change in n

faster rotation



all atoms in upper state

out of phase polarization

in phase polarization

all atoms in lower state

Maximum coherence/polarization for $\pi/2$ pulse

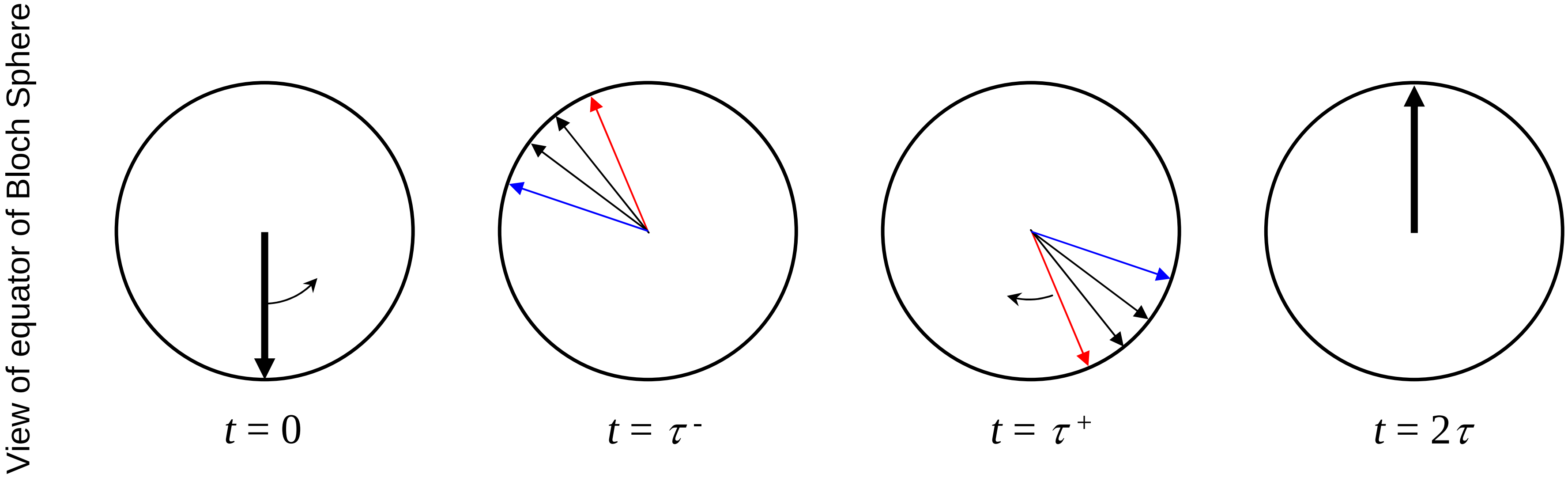
For excitation into off resonance coherence, vector will rotate at $\omega - \omega_0$

Application of Bloch Vector Picture: Photon Echoes

Consider an inhomogeneously broadened system

Excite with a two pulse sequence of a $\pi/2$ pulse followed by a π pulse separated by τ

Each frequency group is described by its own Bloch vector



At $t = 0$ first pulse creates in phase polarization (all Bloch vectors on equator)

Each frequency rotates around axis at own speed, they spread out and the macroscopic polarization disappears

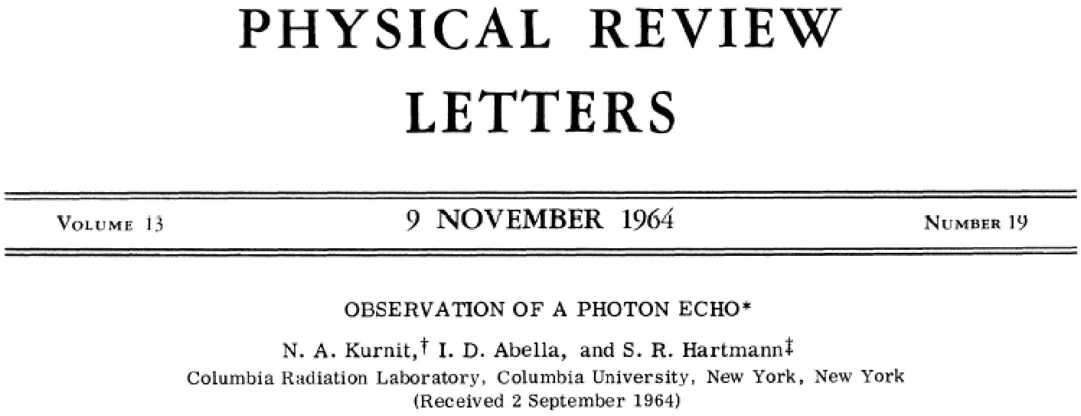
At $t = \tau$, the second pulse flips them over the “north pole” and flips their direction

At $t = 2\tau$, they realign, resulting in a macroscopic polarization, which then radiates a pulse – the photon echo

Photon Echo history

Based on spin echoes observed in magnetic resonance (by Hahn)

First observed in 1964 by Sven Hartmann’s group in ruby



PHYSICAL REVIEW
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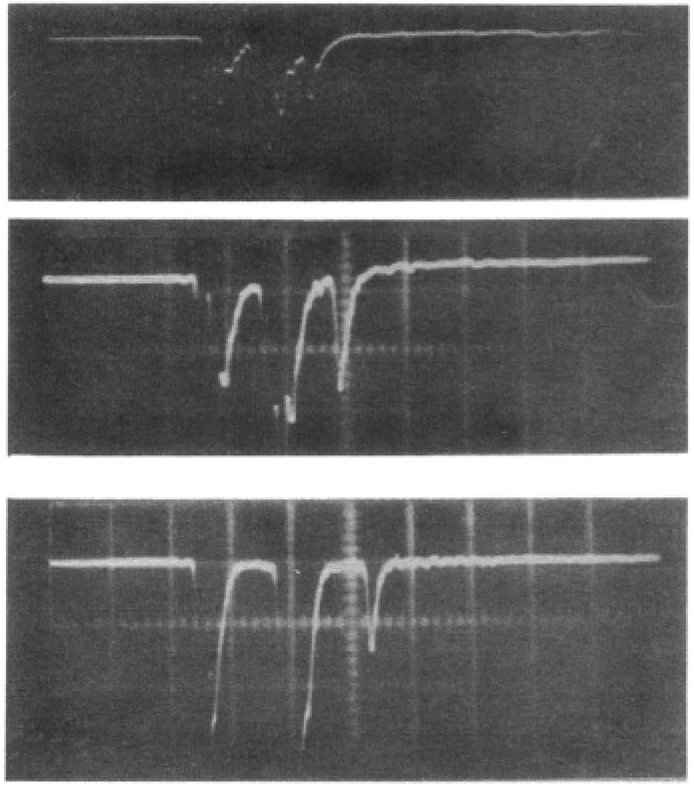
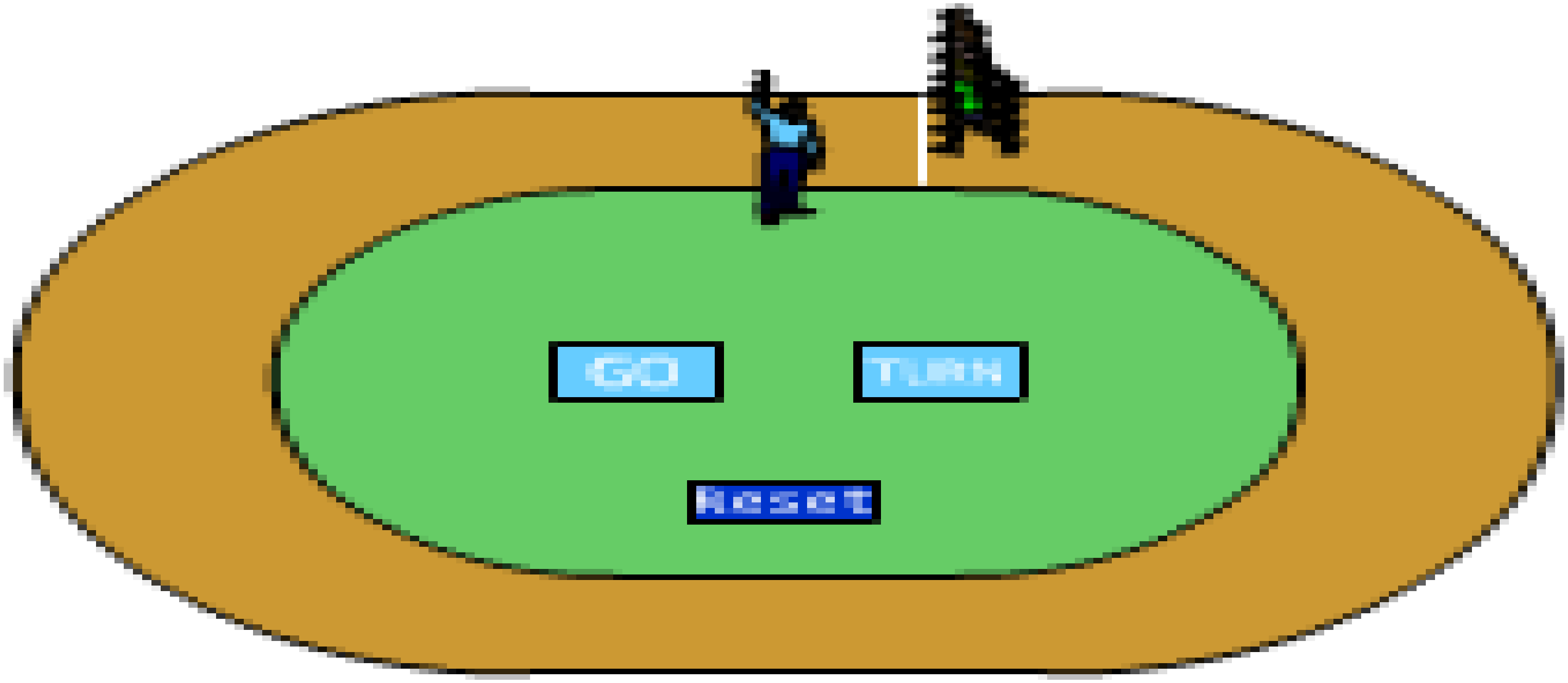
OBSERVATION OF A PHOTON ECHO*

N. A. Kurnit,† I. D. Abella, and S. R. Hartmann‡
Columbia Radiation Laboratory, Columbia University, New York, New York
(Received 2 September 1964)



Although picaresque, the term “photon” echo is misleading as it is not necessary to quantize the electromagnetic field

Layman's Photon Echo



Flash animation by Virginia Lorenz

Perturbation Thoery

Perturbation theory provides analytic solutions to the OBE in the limit of

Weak Fields and

CW

or

Delta function in time ← e.g. short pulses

Outline:

1) First order results: Free decay
    - Homogeneous
    - Inhomogeneous
2) Third order result: Transient four wave mixing
    - Generalization of (a) transient grating or (b) photon echo
        - Measurement of homogeneous linewidth in presence of inhomogeneity
    - Also describes transient absorption
    - Interesting phenomena: spectral diffusion, non-Markovian

Perturbation Theory II

OBEs (not in RWA)

$$\dot{\rho}_{11} = -\gamma_1 \rho_{11} + i\Omega(t)(\rho_{12} - \rho_{21})$$

$$\dot{\rho}_{22} = -\gamma_2 \rho_{22} - i\Omega(t)(\rho_{12} - \rho_{21})$$

$$\dot{\rho}_{12} = -\gamma_{12}^{PH} \rho_{12} + i\omega o\rho_{12} - i\Omega(t)(\rho_{22} - \rho_{11})$$

Expand the OBEs in Ω

$\Omega \to \alpha\Omega$ (α small)

$$\rho_{ij} = \rho_{ij}^{(0)} + \alpha\rho_{ij}^{(1)} + \alpha^2\rho_{ij}^{(2)} + \alpha^3\rho_{ij}^{(3)} + \cdots$$

Insert into OBE and collect coefficients of α^n

$$\dot{\rho}_{11}^{(n)} = -\gamma_1 \rho_{11}^{(n)} + i\Omega(t)\left(\rho_{12}^{(n-1)} - \rho_{21}^{(n-1)}\right)$$

$$\dot{\rho}_{22}^{(n)} = -\gamma_2 \rho_{22}^{(n)} - i\Omega(t)\left(\rho_{12}^{(n-1)} - \rho_{21}^{(n-1)}\right)$$

$$\dot{\rho}_{12}^{(n)} = -\gamma_{12}^{PH} \rho_{12}^{(n)} - i\Omega(t)\left(\rho_{22}^{(n-1)} - \rho_{11}^{(n-1)}\right)$$

Assume $\rho_{11}^{(0)} \neq 0(=1)$, $\rho_{22}^{(0)} = \rho_{12}^{(0)} = 0$

First order:

$$\left.\begin{array}{l} \dot{\rho}_{11}^{(1)} = -\gamma_1 \rho_{11}^{(1)} \\ \dot{\rho}_{22}^{(1)} = -\gamma_2 \rho_{22}^{(1)} \end{array}\right\} \longrightarrow \rho_{11}^{(1)} = \rho_{22}^{(1)} = 0$$

$$\dot{\rho}_{12}^{(1)} = -\gamma_{12}^{PH} \rho_{12}^{(1)} + i\omega_0 \rho_{12}^{(1)} + i\Omega(t)$$

Therefore, to first order the populations don’t change, but a coherence is created

Think of bottom of Bloch sphere, to first order no change vertical, only perpendicular to axis

Perturbation Theory III

Second order:

$$\dot{\rho}_{11}^{(2)} = -\gamma_1 \rho_{11}^{(2)} + i\Omega(t)\left(\rho_{12}^{(1)} - \rho_{21}^{(1)}\right)$$

$$\dot{\rho}_{22}^{(2)} = -\gamma_2 \rho_{22}^{(2)} - i\Omega(t)\left(\rho_{12}^{(1)} - \rho_{21}^{(1)}\right)$$

$$\dot{\rho}_{12}^{(2)} = -\gamma_{12}^{PH} \rho_{12}^{(2)} + i\omega_0 \rho_{12}^{(2)} - i\Omega(t)\underbrace{\left(\rho_{22}^{(1)} - \rho_{11}^{(1)}\right)}_{=0} \quad \Rightarrow \quad \rho_{12}^{(2)} = 0$$

This shows that at second order, populations are generated, but the coherence is unchanged.

This means that populations are proportional to E^2, consistent with number of atoms in excited state being related to intensity.

It is instructive to draw the following diagram:

$$\begin{tikzcd}
 & \rho_{12}^{(1)} \arrow[r] \arrow[rdd] & \rho_{11}^{(2)} \arrow[r] \arrow[rdd] & \rho_{12}^{(3)} \\
\rho_{11}^{(0)} \arrow[ru] \arrow[rd] & & & \\
 & \rho_{21}^{(1)} \arrow[r] \arrow[ruu] & \rho_{22}^{(2)} \arrow[r] \arrow[ruu] & \rho_{21}^{(3)}
\end{tikzcd}$$

From this, we conclude new contributions to the polarization arise at odd orders, whereas new contributions to the population arise at even orders

First order (linear) solution

To first order, only the off-diagonal element changed

$$\dot{\rho}_{12}^{(1)} = -\gamma_{12}^{PH}\rho_{12}^{(1)} + i\omega_0\rho_{12}^{(1)} + i\Omega(t)$$

$$\dot{\rho}_{12} + \left(\gamma_{12}^{PH} - i\omega_0\right)\rho_{12} = i\Omega(t)$$

$$\left[\dot{\rho}_{12} + \left(\gamma_{12}^{PH} - i\omega_0\right)\rho_{12}\right]e^{(\gamma_{PH} - i\omega_0)t} = e^{\left(\gamma_{12}^{PH} - i\omega_0\right)t}\Omega(t)$$

$$\frac{d}{dt}\left[\rho_{12}e^{\left(\gamma_{12}^{PH} - i\omega_0\right)t}\right] = e^{(\gamma_{PH} - i\omega_0)t}\Omega(t)$$

$$\rho_{12}e^{\left(\gamma_{12}^{PH} - i\omega_0\right)t} = \int_{-\infty}^{t} e^{\left(\gamma_{12}^{PH} - i\omega_0\right)t'}\Omega(t')dt'$$

$$\boxed{\rho_{12} = i\int_{-\infty}^{t} e^{\left(-\gamma_{12}^{PH} + i\omega_0\right)(t-t')}\Omega(t')dt'}$$

This integral can be easily done in two cases

1) CW field – we basically did this in analyzing RWA

2) Infinitely short pulse: Free $\left\{\begin{array}{l}\text{Induction}\\ \text{Polarization}\end{array}\right\}$ Decay

Homogeneous Free Decay

Write $\Omega(t) = \hat{E}(t)e^{i\omega t} + \hat{E}^*(t)e^{-i\omega t}$ and let $\rho_{12} = \hat{\rho}_{12}e^{i\omega t}$

$$\hat{\rho}_{12} = e^{-i\omega t} i \int_{-\infty}^{t} e^{\left(-\gamma_{12}^{PH}+i\omega_0\right)(t-t')} \left(\hat{E}(t')e^{i\omega t'} + \cancel{\hat{E}^*(t')e^{-i\omega t'}}\right) dt'$$

Drop by RWA

$$\hat{\rho}_{12} = e^{-i\omega t} i \int_{-\infty}^{t} e^{\left(-\gamma_{12}^{PH}+i\Delta\omega\right)(t-t')} \hat{E}(t')e^{i\omega t'} dt' \qquad (\Delta\omega = \omega_0 - \omega)$$

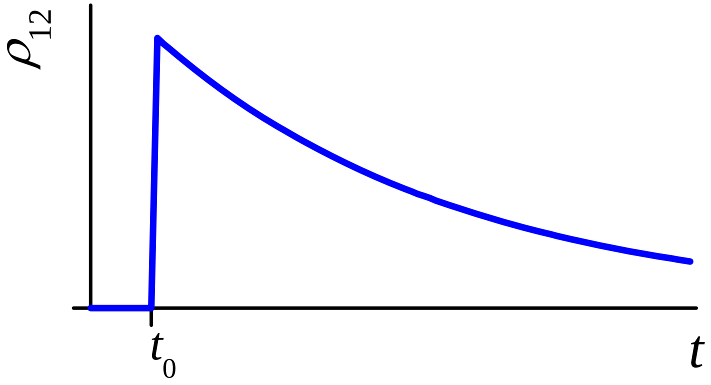
Let $\hat{E}(t) = \delta(t - t_0)$ and do integral

$$\hat{\rho}_{12}(t) = ie^{\left(-\gamma_{12}^{PH}+i\Delta\omega\right)(t-t_0)}\Theta(t-t_0)$$

($\Theta(t - t_0)$ is step function)

But $\hat{\rho}_{12} = \rho_{12}e^{-i\omega t}$

$$\rho_{12}(t) = ie^{-i\omega_0 t}e^{-\gamma_{12}^{PH} t}e^{-i\Delta\omega t_0}\Theta(t-t_0)$$



This a field that oscillates at the resonance frequency, with a phase determined by laser and decays exponentially, starting at $t = t_0$

Note: time domain decay at rate γ, frequency linewidth is $\pi\gamma$

Free decay = absorption

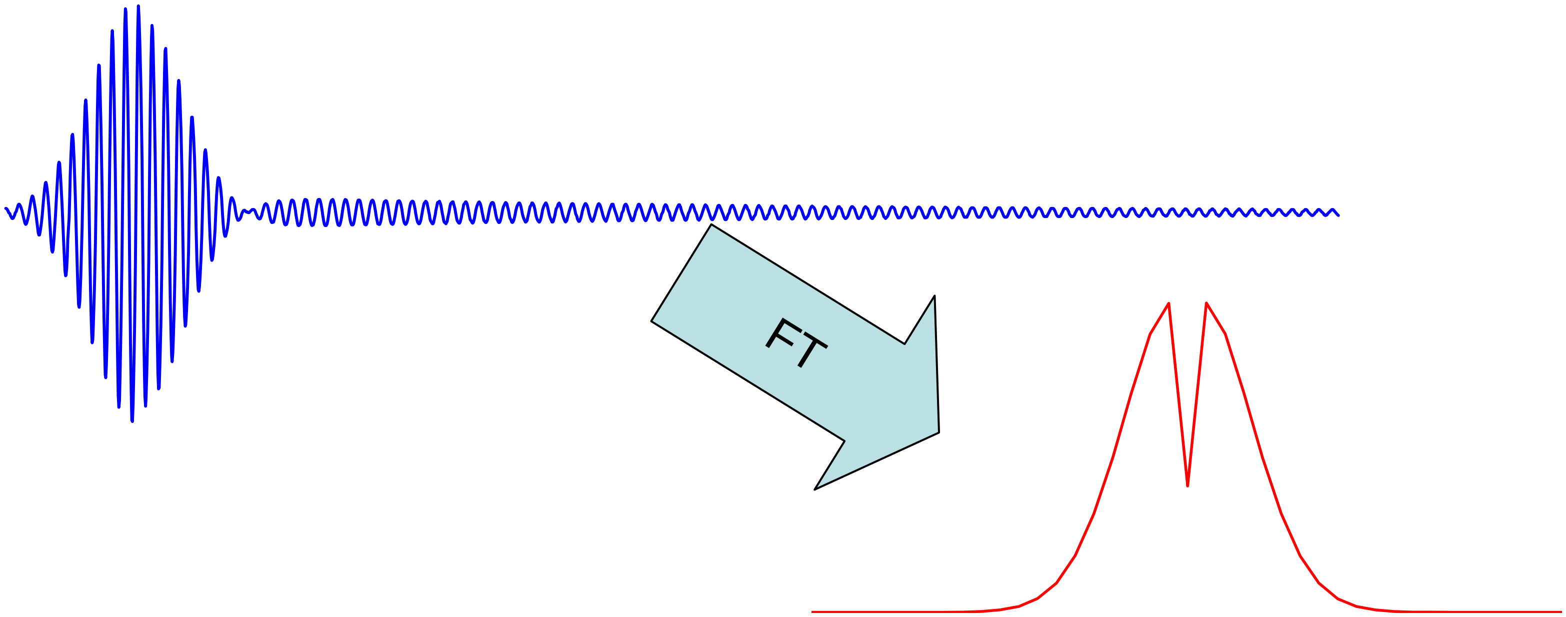
This free decay is nothing but the polarization that is responsible for absorption, there is one factor of i in ρ_{12}

$$\rho_{12}(t) = ie^{-i\omega_0 t} e^{-\gamma_{12}^{PH} t} e^{-i\Delta\omega t_0} \Theta(t - t_0)$$

and another i comes from Maxwell’s equations (recall lecture 2) to make the reradiated field 180° out of phase with the incident pulse

this means that the reradiated field destructively interferes with the incident pulse

….huh? …they don’t overlap in time… yes but it is still correct, you can think of the destructive interference in the frequency domain if it helps



Inhomogeneous Free Decay

Integrate homogeneous result over inhomogeneous distribution

$$g(\omega_0) = \frac{1}{\sqrt{\pi\alpha^2}} \exp\left[-(\omega_0 - \omega_c)^2 / \alpha^2\right] \qquad \text{where} \quad \alpha^2 = \frac{(\delta\omega)^2}{4\ln 2}$$

$\delta\omega$ is the FWHM of the distribution centered at ω_c

for simplicity, let t_0 → 0

$$\rho_{12}^T = \int_{-\infty}^{\infty} \rho_{12}(\omega_0) g(\omega_0) d\omega_0 = \underbrace{\frac{i\Theta(t) e^{-\gamma_{12}^{ph} t}}{\sqrt{\pi\alpha^2}}}_{A} \int_{-\infty}^{\infty} e^{-i\omega_0 t} e^{-(\omega-\omega_0)^2/\alpha^2} d\omega_0$$

$$= A \int_{-\infty}^{\infty} \exp\left[-\left(\omega_0^2 - \left(2\omega_c - i\alpha^2 t\right)\omega_0 + \omega_0^2\right)/\alpha^2\right] d\omega_0$$

$$\vdots$$

$$= i\Theta(t) e^{-\gamma_{12}^{ph} t} e^{-i\omega_c t} e^{-\frac{\alpha^2}{4} t^2}$$

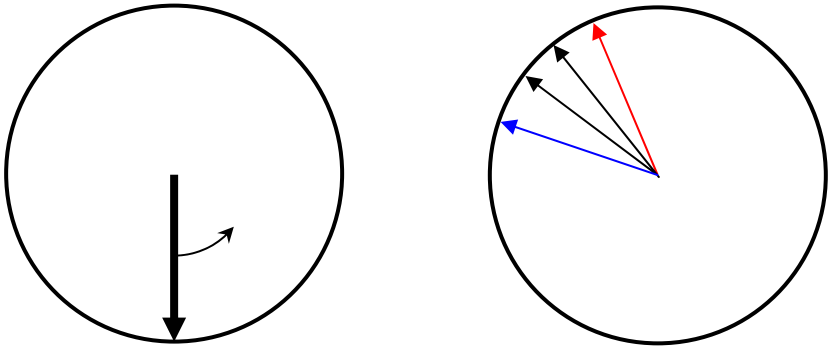
this is a field that oscillates at the center frequency of the inhomogeneous distribution, but it decays like a Gaussian with a time constant determined by the inhomogeneous width (assuming it is larger than the homogeneous width)

Inhomogeneous Free Decay and Absorption

Inhomogeneous free decay is due to interference among different frequency components.

Similar to destructive interference between frequencies that produces a pulse

Corresponds to spreading of Bloch vectors



$t = 0$ $t = \tau^-$

Again this polarization is responsible for the linear absorption, with the faster decay corresponding to the broader absorption line due to inhomogeneous broadening

Third order perturbation theory → Four wave mixing

Taking perturbation theory to 3rd order addresses wave-mixing spectroscopy

- Most common is four-wave-mixing (FWM)

FWM mixing can be done in

- time domain (changing delays)
- frequency domain (changing relative detunings)
- in both cases, there is also an overall tuning with respect to the resonance

Time-domain version (Transient FWM or TFWM) encompasses

- Photon-echoes
- Transient grating
- Transient absorption

Frequency-domain version encompasses

- Self-phase modulation
- Spectral broadening

Four wave mixing geometries I

Consider generic 3rd order nonlinear process

$$P^{(3)}(\omega_4,\mathbf{k}_4,t)=\chi^{(3)}(\omega_1,\omega_2,\omega_3,\omega_4)E_1(\omega_1,\mathbf{k}_1,t)E_2(\omega_2,\mathbf{k}_2,t)E_3(\omega_3,\mathbf{k}_3,t)$$

We need perturbation theory to calculate $\chi^{(3)}$

Note: all fields on right side do not need to be unique

There are two conservation laws

(we will consider cases were the ω_i's are similar)

$$\omega_4=\omega_i+\omega_j-\omega_k \qquad \mathbf{k}_4=\mathbf{k}_i+\mathbf{k}_j-\mathbf{k}_k$$

perturbation theory will tell us which field(s) can have the minus sign

Phase matching

the addition of $\mathbf{k}$'s is often called “phase matching”

also need to consider if $P^{(3)}$ stays in phase with the radiated field during propagation through the sample

more significant for thicker samples

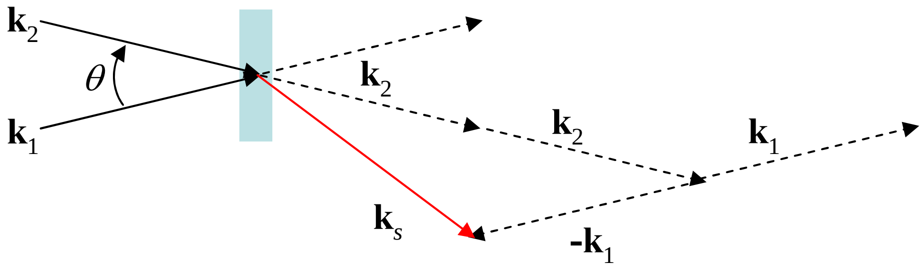
Four wave mixing geometries II

Self-diffraction: grating forms between pulses, then second pulse scatters off it

Only two fields incident

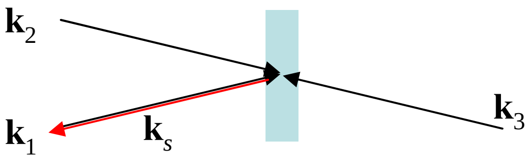
$$\mathbf{k}_s = 2\mathbf{k}_2 - \mathbf{k}_1 \qquad \omega_s = 2\omega_2 - \omega_1$$

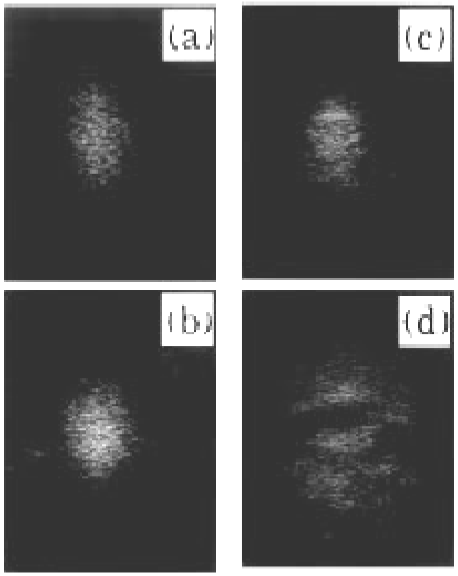
quasi phase-matched:
phase matched for small θ
small phase mis-match okay
for thin samples



Phase conjugate

$$\mathbf{k}_s = \mathbf{k}_3 + \mathbf{k}_2 - \mathbf{k}_1 = -\mathbf{k}_1 \qquad \omega_s = \omega_3 + \omega_2 - \omega_1$$





Exactly phase matched
works in thick samples

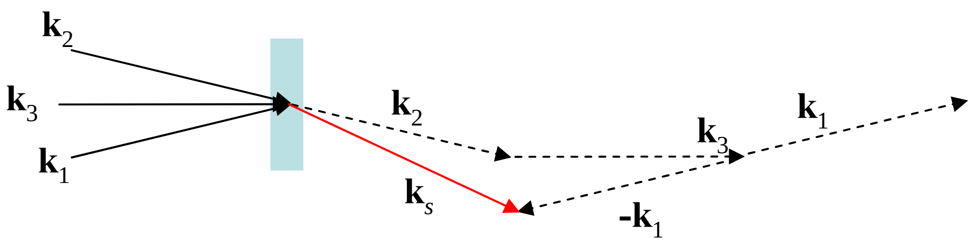
Four wave mixing geometries III

Three pulse (same as transient grating)

$$\mathbf{k}_s = \mathbf{k}_3 + \mathbf{k}_2 - \mathbf{k}_1 = \qquad \omega_s = \omega_3 + \omega_2 - \omega_1$$

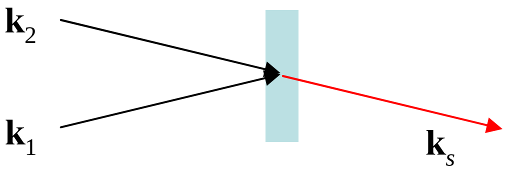
quasi phase-matched:

phase matched for small angle
small phase mis-match okay
for thin samples



Pump-probe (transient absorption)

$$\mathbf{k}_s = \mathbf{k}_2 + \mathbf{k}_1 - \mathbf{k}_1 = \mathbf{k}_2$$



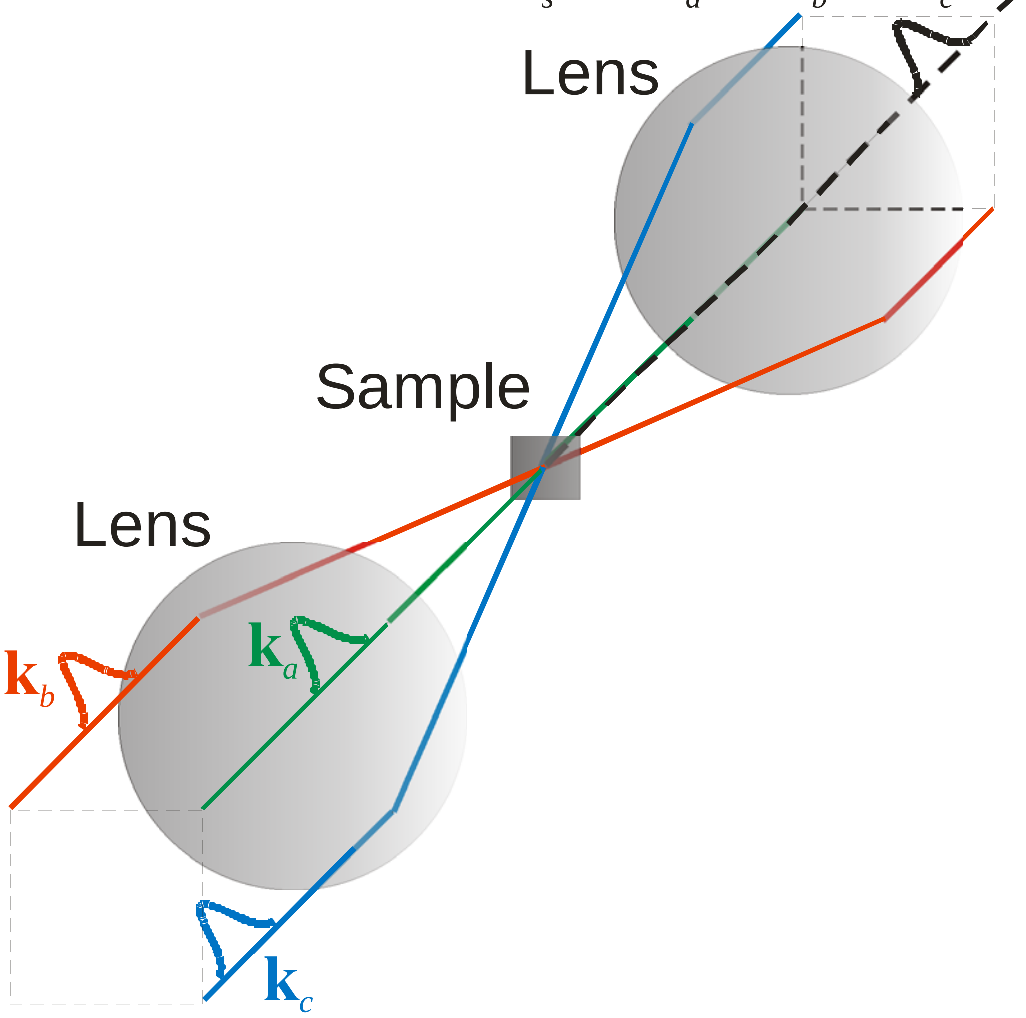
Third order correction to the free decay (which is absorption….)

Four wave mixing geometries IV

Box Geometry (phase matched, needs minimum clear aperture)

$$\mathbf{k}_s = \mathbf{k}_3 + \mathbf{k}_2 - \mathbf{k}_1 = \qquad \omega_s = \omega_3 + \omega_2 - \omega_1$$

$$\mathbf{k}_s = -\mathbf{k}_a + \mathbf{k}_b + \mathbf{k}_c$$



Perturbation calculation for self diffraction I

Do expansion as outline before, plug lower orders into higher orders

Each order is integral over the next lower order

$$\rho_{12}^{(3)}(t) = \frac{i\mu_{12}^3}{\hbar^3} \int_{-\infty}^{t} E(t_3)\exp\left[\left(i\omega_0 - \gamma_{ph}\right)(t-t_3)\right]$$

$$\times \int_{-\infty}^{t_3}\int_{-\infty}^{t_2} [E(t_2)E(t_1)(\exp[-\gamma_2(t_3-t_2)] + \exp[-\gamma_1(t_3-t_2)])\exp\left[\left(i\omega_0 - \gamma_{ph}\right)(t_2-t_1)\right] + c.c.]dt_1dt_2dt_3$$

This result is general, we haven assumed anything about the field or made any approximations.

Now write $E(t) = \hat{E}(t)\exp(i\omega t) + \hat{E}^*(t)\exp(-i\omega t)$ and $\rho_{12}^{(3)} = \hat{\rho}_{12}^{(3)}\exp[i\omega t]$ $\Delta\omega = \omega_0 - \omega$

$$\rho_{12}^{(3)}(t) = \frac{i\mu_{12}^3}{\hbar^3} \int_{-\infty}^{t} \hat{E}(t_3)\exp\left[\left(i\Delta\omega - \gamma_{ph}\right)(t-t_3)\right]$$

$$\int_{-\infty}^{t_3}\int_{-\infty}^{t_2} \left[\hat{E}^*(t_2)\hat{E}(t_1)(\exp[-\gamma_2(t_3-t_2)] + \exp[-\gamma_1(t_3-t_2)])\right]\exp\left[\left(i\omega_0 - \gamma_{ph}\right)(t_2-t_1)\right] + c.c.]dt_1dt_2dt_3$$

Perturbation calculation for self-diffraction II

Now write $\hat{E}(t)=a_1e(t)\exp[-ik_1r]+a_2e(t-\tau)\exp[-ik_2r]$

and choose terms in $2\mathbf{k}_2$-$\mathbf{k}_1$ direction

$$
{}_{2k_2-k_1}\hat{\rho}_{12}^{(3)}=\frac{i\mu_{12}^3}{\hbar^3}\int_{-\infty}^{t}a_2e(t_3-\tau)\exp[(i\Delta\omega-\gamma_{ph})(t-t_3)]
$$

$$
\times\int\int\left[a_1^*e(t_2)a_2(t_1-\tau)\exp[(i\Delta\omega-\gamma_{ph})(t_2-t_1)]+a_1^*e(t_1)a_2(t_2-\tau)\exp[(i\Delta\omega-\gamma_{ph})(t_2-t_1)]\right]
$$

$$
\times(\exp[-\gamma_2(t_3-t_2)]+\exp[-\gamma_1(t_3-t_2)])dt_1dt_2dt_3
$$

let $e(t)$ → $\delta(t)$ and do integrals

$$
{}_{2k_2-k_1}\hat{\rho}_{12}^{(3)}=\frac{i\mu_{12}^3}{\hbar^3}a_2a_2a_1^*\Theta(t-\tau)\Theta(\tau)\exp[(i\Delta\omega-\gamma_{ph})(t-\tau)]\exp[(-i\Delta\omega-\gamma_{ph})\tau]
$$

$$
=\frac{i\mu_{12}^3}{\hbar^3}a_2a_2a_1^*\exp[(i\Delta\omega-\gamma_{ph})t-2i\Delta\omega\tau]
$$

We see:

- only one term [other dropped because of $\Theta(\tau)\Theta(-\tau)$]
- need $\tau>0,\ t>\tau$
- signal decays in real time with γ_{ph}, oscillates at ω_0