

Two paths may interfere as they are inside the cell.

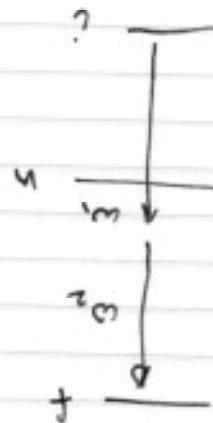
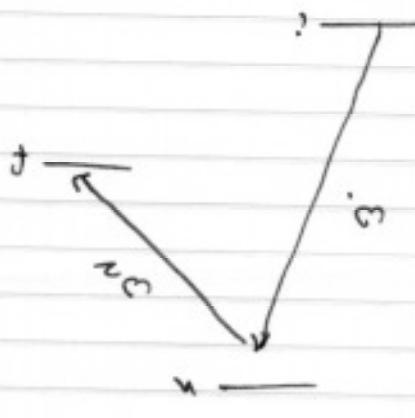
by  $\omega_2$ , and vice-versa.

two terms correspond to absorption of  $\omega_1$  follow

$$\left| \frac{\omega_3 - \omega_2}{\epsilon(1/3 \cdot P) \ln(1/3 \cdot P)} \right| \geq \left| \frac{\omega_3 - \omega_1}{\epsilon(1/3 \cdot P) \ln(1/3 \cdot P)} \right| \leq \left| \frac{\omega_3 + \omega_1}{\epsilon(1/3 \cdot P) \ln(1/3 \cdot P)} \right| \approx \frac{g_{-} \epsilon}{P}$$

2nd-order perturbation theory  
from M. Gippert-Mayer

Two-photon absorption transitions



Two-photon transitions transitions

①

gives Doppler shift ( $\omega_1 \neq \omega_2$ ).

a factor  $\frac{k_1 - k_2}{k_1 + k_2}$ , which can be

the Doppler shift can be reduced by

Clearly if we choose  $k_1 = -k_2$

Doppler-shifted frequency

$$\omega_3 = \omega_1 - \omega_2 = \omega_1 + \omega_2 - 2(k_1 + k_2)$$

Since the resonance condition should be

$$\omega_3 = k_1 + k_2$$

$$\left( \omega_1 - \frac{\omega_1 + \omega_2}{2} k_1 + \omega_2 + \frac{\omega_1 + \omega_2}{2} k_2 \right) + \left( \omega_1 + \omega_2 - \frac{\omega_1 + \omega_2}{2} k_1 - \frac{\omega_1 + \omega_2}{2} k_2 \right) \times$$

$$= \frac{[\omega_1 + \omega_2 - \omega_1 - \omega_2] + (\omega_1 + \omega_2)}{\omega_1 + \omega_2}$$

$$P_{\text{eff}} \propto I^1 I^2$$

now consider other wise, and include Doppler shift

(2)

$$\left| \frac{\omega_1 - \omega_2}{(k_1^2 + k_2^2 + k_3^2) + (\omega_1^2 + \omega_2^2)} \right| > \frac{(\omega_1^2 - 2\omega_1 - 2k_1^2)^2 + (\omega_2^2 - 2\omega_2 - 2k_2^2)^2}{\omega_1^2 + \omega_2^2}$$

$$\left| \frac{\omega_1 - \omega_2}{(k_1^2 + k_2^2 + k_3^2) + (\omega_1^2 + \omega_2^2)} \right| > \frac{(\omega_1^2 - 2\omega_1 + 2k_1^2)^2 + (\omega_2^2 - 2\omega_2 + 2k_2^2)^2}{\omega_1^2 + \omega_2^2}$$

$$P_{\text{tot}} = I_1 I_2 \frac{\omega_1 - \omega_2}{\omega_1^2 + \omega_2^2} \frac{(\omega_1^2 - 2\omega_1)^2 + (\omega_2^2 - 2\omega_2)^2}{(k_1^2 + k_2^2 + (\omega_1^2 - 2\omega_1)(\omega_2^2 - 2\omega_2))^2}$$

In the case where  $\omega_1 = \omega_2$ ,  $k_1 = -k_2$

or if  $k_1 \neq k_2$ , as well as 2 form  $k_1$  or 2 form

can be 2-photon contributions from absorption

For a single frequency,  $k_1 = k_2$ , there

number.

neglected, since  $\omega_1 - \omega_2$  is generally a large

of the 2nd-order matrix elements can be

-generally the Doppler-term in the denominators

(3)

for example  
observed is  $1 + 15 \rightarrow 25$  transistors

Note: although only 1 pt. in 10", have been

( $\frac{1}{2} \sqrt{\frac{C}{C_0}} \omega_0^2$ )  
4) we have ignored quadratic Doppler shifts

(determining is large compared to Rayleigh frequency)

$$3) \Delta \omega_n - \omega_0 \ll \left( \frac{1}{2} \sqrt{\frac{C}{C_0}} \omega_0 \right)^2$$

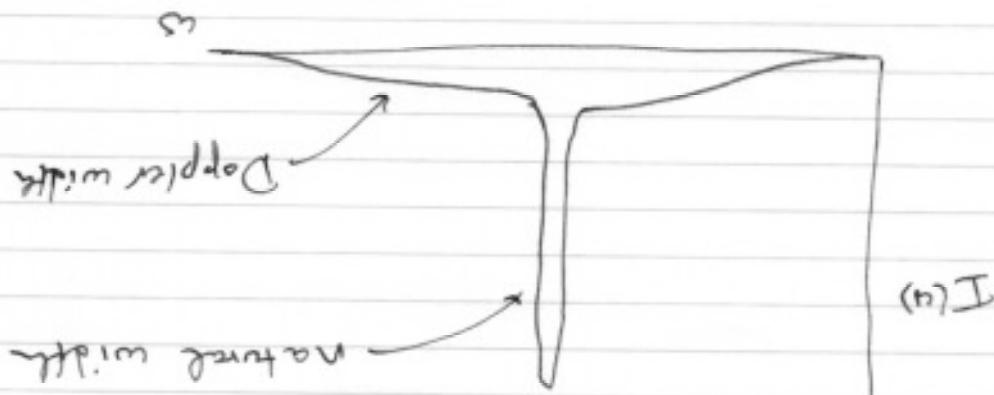
$$|\omega_{nq} - \omega_0| \ll k_B \text{ and } |\omega_{nq} - \omega_0| \ll \gamma_n$$

2) intermediate states are off-resonant

1)  $\gamma_1 \neq \gamma_2$  are non-degenerate

and regular

Note: these expressions are perturbative



The lineshape will look like

④

is not allowed for  $S \rightarrow S$ . That's why there is no absorption of 2 photons from a single beam.



eliminate Doppler background:

for  $S \leftarrow S$  transitions, it is possible to

(and opposite for  $|g\rangle$ )

Parity must be the same for  $|g\rangle$  &  $|e\rangle$

where  $g = -1$  for  $\text{D}^-$ , 0 for  $\text{D}^+$ , +1 for  $\text{D}^+$

$$\text{and } \Delta m_F = g_1 + g_2$$

$$\text{we find } |\Delta F| = 2$$

so E1 rules apply.

mixing elements are still products of dipole operators,

The intermediate states are far-off resonance, the

a pair of E1 transitions (note: even if

Two-photon selection rules

⑤

$|e\rangle$  is a ground state with  $\sim 0$  width.  
 important for Raman transitions, where  
 the same is true for  $|g\rangle$ , which may be

$$|g\rangle \propto I_1(\omega_1 d \cdot \beta, i\gamma)^2$$

ground state

This produces an effective linewidth of the  
 laser a linewidth of  $\gamma_{bg}$ .

state gets an admixture of the state  $|u\rangle$ , which  
 results from dressed atom model, the ground

$$\Delta E_g \propto I_1(\omega_1 d \cdot \beta, i\gamma)^2$$

the ground state shifts

using 2nd-order perturbation theory.

Lights shifts : linewidths

⑥

2-photon resonance.

a light shift as one scans through

for a two-photon transition, there can be

(as in resonance, changes sign red or blue)

light shift is net observable

Note: for a single-photon transition, the

$$\frac{(\gamma/m) + (\gamma_m - \kappa^2 - \omega^2)}{\kappa^2} |C|^2 + I^2$$

$$\Delta g = I_1 |C|^2 - I_2 |C|^2$$

and a similar expression for the total linewidth

$$-\frac{(\gamma/m) + (\gamma_m - \kappa^2 - \omega^2)}{\kappa^2} |C|^2 - I^2$$

$$\Delta E_g = I_1 |C|^2 - I_2 |C|^2$$

shift as well, so that

there is of course a light shift of the final

②

- can be Doppler free if  $\Delta k = 0$

- will scale as  $I^n$

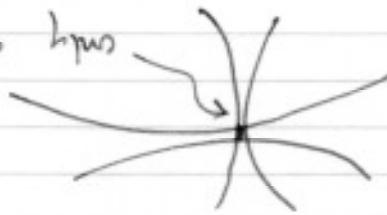
absorption

- one can generalize to n-photon

occurs.

z-photon transitions

only at intersection can



with crossed beams:

- one can also gain spatial selectivity

part of laser beam.

absorption is concentrated in very intense

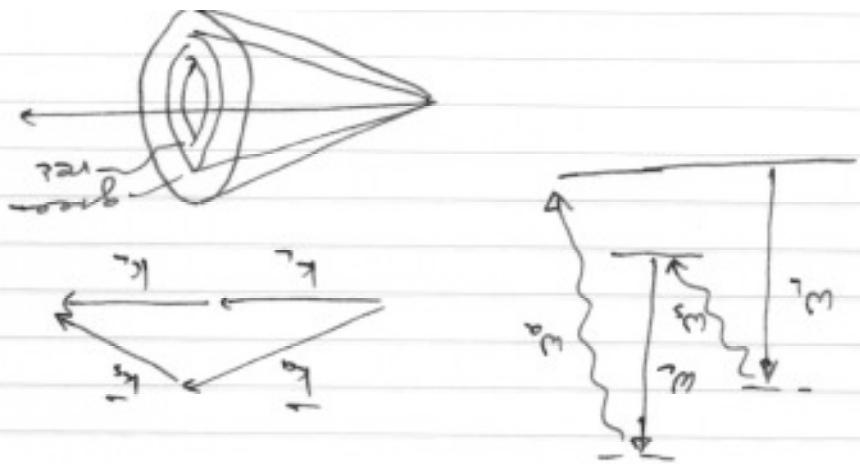
useful in 2-photon microscopy, since

$\Leftrightarrow$  nonlinear transistion

Transitions scales as  $I, I^2$  (or  $I^2$  for single color).

some points about 2-photon spectroscopy

⑧

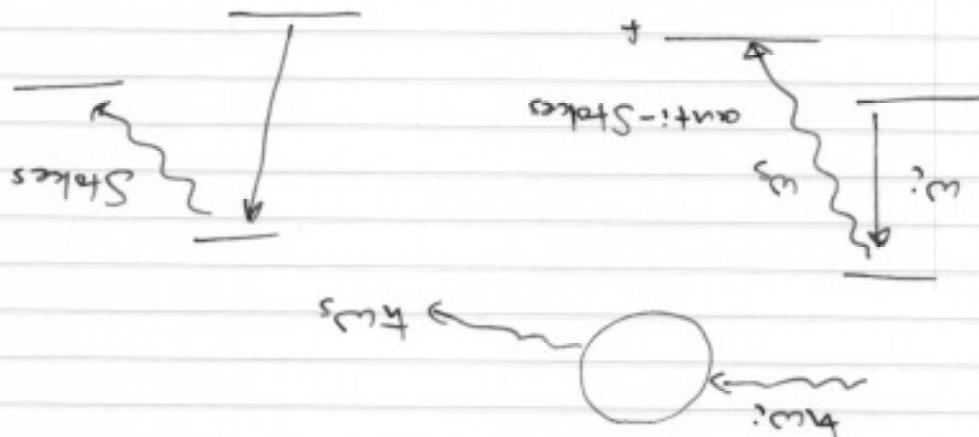


higher-order processes (shimulated Larmor)

Leads of a molecule  
• rotation

Typically  $E^i, E^f$  are different vibrational

energy conservation:  $\hbar\omega_s - \hbar\omega_i = E^i - E^f$



- Larmor scattering:

- used for molecular transistors

Larmor spectroscopy

(6)

$$|\Psi(t=0)\rangle = C_1 |\Psi_1\rangle + C_2 |\Psi_2\rangle$$

if we consider two states, initially

$$\text{area pulse width} \rightarrow \sim \frac{1}{\Delta \omega} < \frac{\hbar}{\Delta E_{\text{tot}}}$$

$$\Delta \omega > \frac{\hbar}{E_{\text{tot}} - E_{\text{tot}}^*}$$

- This will require an oscillation bandwidth

different energies.

which is time-dependent, since the  $|\Psi_t\rangle$  have

$$|\Psi(t)\rangle = \sum^* C_k |\Psi_k\rangle e^{-iE_k t/\hbar}$$

forming a superposition state

to simultaneously excite a number of states,

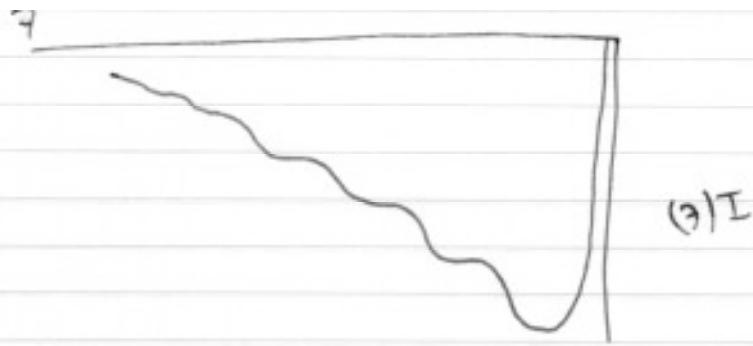
manifold, for example), it may be possible

$|\Psi_t\rangle$  describe closely spaced states (upper fine

$$|\Psi_t\rangle = E_{\text{tot}} |\Psi_t\rangle$$

consister atomic states  $|\Psi_t\rangle$

Quantum Beat Spectroscopy



This yields

$$B = 2C_1 C_2 |\langle \psi_1 | \tilde{J} | \psi_2 \rangle| < 4 |\tilde{J}|^2 |\psi_1|^2 |\psi_2|^2$$

$$A = |C_1|^2 |\tilde{J}|^2 |\psi_1|^2 + C_2^2 |\tilde{J}|^2 |\psi_2|^2$$

where  $\tilde{J} = J_1 - J_2$   $\omega_1 = \omega_2 - \omega_0$

$$S = C e^{-\tilde{J}t} (A + B \cos \omega_1 t)$$

using form for  $A(\epsilon)$  yields

where  $\tilde{J}$  is the polarization vector of the emitted light

$$S(t) \propto C |\langle \psi_1 | \tilde{J} | \psi_2(t) \rangle|^2$$

from both levels. It will see a signal

Imagine a detector that will measure Fluorescence

where  $C_0$  is enough difference  $\omega_1 - \omega_0$

$$\psi_2(t) = \sum_k C_k e^{i k \omega_0 t} (C_{k0} + F_{k0})$$

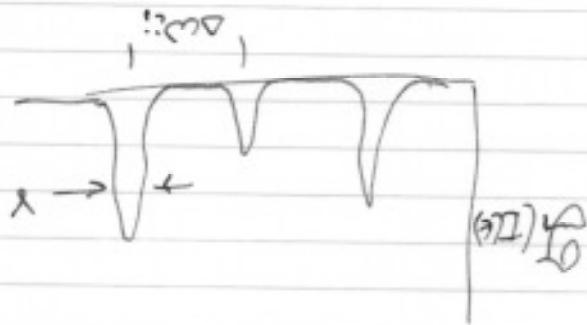
with decay widths  $\lambda_k$ , then

if we assume  $|4_1\rangle$ ;  $|4_2\rangle$  are excited states



case between internal states of atoms.  
— this is an example of coherence, in this

through the laser bandwidth is  $\Delta\nu$ , even  
— note this gives a spectral resolution of  $\delta$ ,



using Fourier transform  
more complicated. Typically find  $\omega_0$  by  
— If one oscillates multiple levels, the beating is

excited state hyperfine structure.  
Quantum beats are a good way to measure

is enhanced by  $N$  due to coherence.

⇒ example of super-redundance, where emission

$$\text{then } \left| \sum_{i=1}^N d_{i2} \right|^2 = \left| N d_{12} \right|^2 = N^2 |d_{12}|^2 = N \cdot I_{12}$$

occurred for a short laser pulse,

if the algorithm is coherent, as usual we

$$\text{and } I_{12} = N \text{ tru } A_{21}$$

$$\left| \sum_{i=1}^N d_{i2} \right|^2 = \left| \sum_{i=1}^N d_{12} \right|^2$$

no phase relation between atoms, and

- if algorithm is incoherent, then there is

↳ different memory elements

$$I = \sum_{i=1}^N \text{tru } A_{21} = \frac{e^{-\omega_2 t}}{\epsilon^2 \omega^3} \sum_{i=1}^N |d_{12}|^2$$

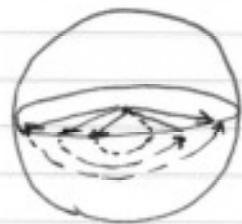
then the total fluorescence intensity is

to solve 127

- assume  $N$  atoms are coherent simultaneously

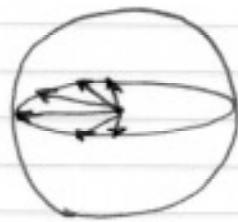
Photon Echoes

(13)

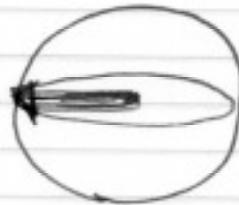


4) apply  $\pi$  pulse

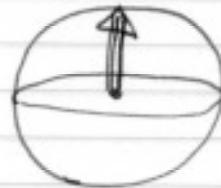
Individual Block vectors  
rotate differently  
due to different environment



3) let system evolve for time  $\tau$



2) apply  $\pi/2$  pulse (put 50% of atoms in |2>)



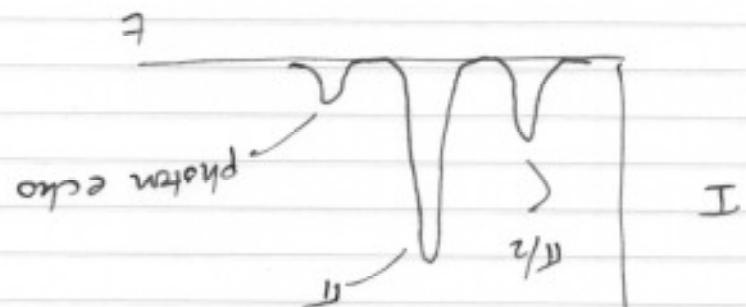
1) initial

$N$  atoms that are inhomogeneously readout.

- consider evolution of Block vectors for

(4)

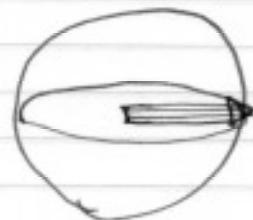
- another example of Doppler-free spectroscopy



If would be at  $t = 2\tau$  without  $\pi$ -pulse  
emission is  $N^2$  times larger than

super-rebunch emission.

- since these atoms are all in phase, get



5) allow system to evolve to  $t = 2\tau$

and no Doppler background.  
resolution of order the natural linewidth,

Show that the spectrum recorded has a

$$\text{an output } A = \frac{1}{\pi} \int_{-\infty}^{\infty} \cos(2\pi(f_1 + f_2)t) \cdot S(t) dt.$$

a lock-in amplifier at  $f_1 + f_2$ . [This gives

B) The fluorescence signal is detected with

for the signal  $S(t, \Delta)$   
proportional to  $\Delta N \cdot I$ , write an expression

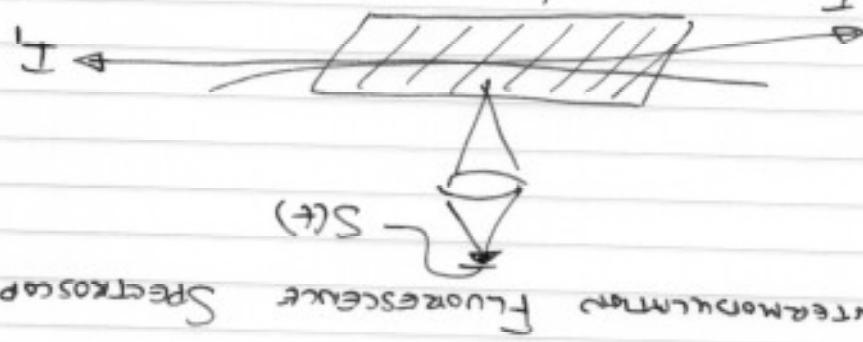
A) Using the fact that fluorescence is

$$k_1 = -k_2 \text{ (counter-propagating beams)}$$

$$I_2 = I_0 + I' \cos 2\pi f_2 t$$

$$I_1 = I_0 + I' \cos 2\pi f_1 t$$

$N$  atoms, doppler width  $\Delta$



Homework Due 10/16