5- Lasers and spectroscopy

Lasers: • coherent and monochromatic light

• linewidth $\leq$ linewidth of the system to be investigated

$\Rightarrow$ Allow studies of much higher resolution than that obtained with conventional spectrometers

5.(a) - Hole burning

(+) Key idea: a strong signal applied to a transition causes the population difference on that transition to decrease

$\Rightarrow$ Decrease in the laser - intensity profile

• Physics behind it: The intensity of the radiation in a cavity cannot increase indefinitely, because stimulated transitions start to reduce the inversion density below the value it had in the absence of oscillations

• Steady state: gain at oscillation frequency $\equiv$ cavity losses

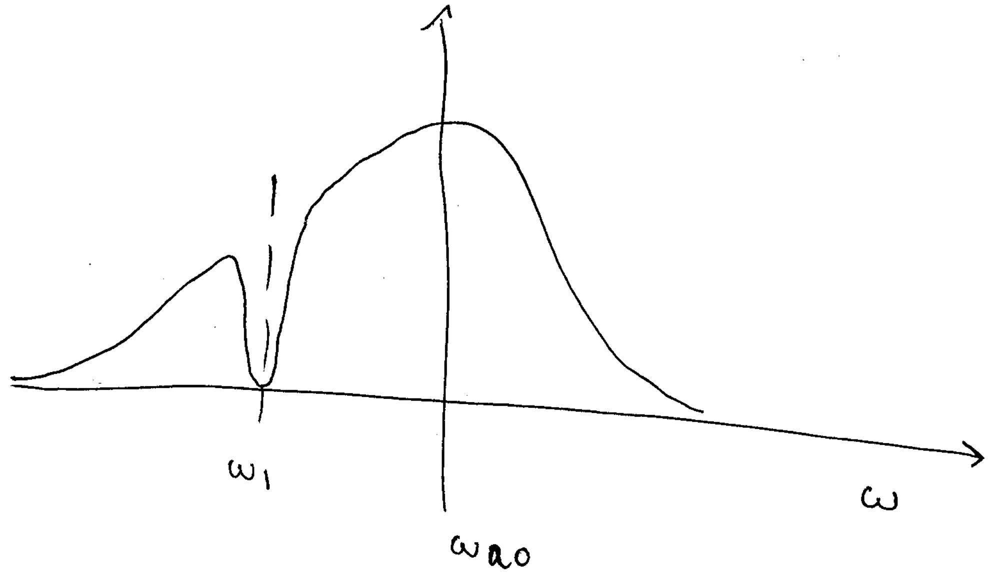
$\Rightarrow$ Gain saturation

Hole burning occurs in inhomogeneously broadened transitions (groups of atoms with slight different resonant frequencies have different saturation properties).

Example: Inhomogeneous, Doppler broadened gas lasers
Inhomogeneous

• Nuclear magnetic resonance transitions (MICROWAVE)

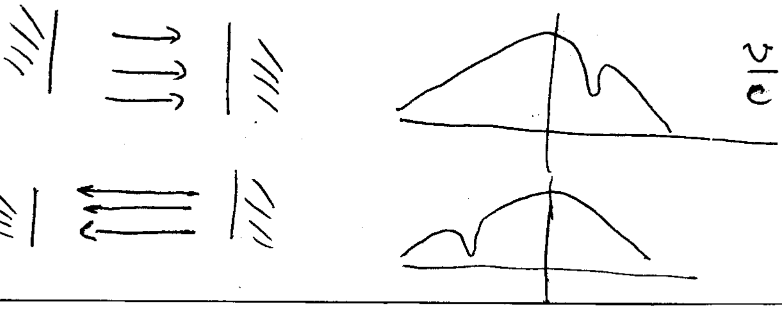
<u>Hole burning</u> : A strong applied signal at frequency ω_1 will saturate the population difference only for those atoms which are nearly resonant with ω_1. The remaining atoms will remain unchanged



<u>Example</u> : Lamb dip (W. E. Lamb, Phys. Rev. 134, 1429(1964))

- "If one tunes the resonance frequency of a single oscillating cavity across a Doppler-broadened gas laser transition, the curve of oscillation shows a narrow dip when the oscillation frequency coincides with the center of the laser-broadened line"

⇒ Hole burning caused by the two opposite waves traveling in the cavity



$$\frac{v}{c} = \frac{\omega - \omega_0}{\omega_0} \; ; \; \omega = \omega_0 + \omega_0 \frac{v}{c}$$

$$\frac{v}{c} = \frac{-(\omega - \omega_0)}{\omega} \; ; \omega = \omega_0 \left(1 - \frac{v}{c}\right)$$

(*) Elementary analysis (based on susceptibility changes)

Let us consider the susceptibility $\tilde{\chi}_n(\omega, \omega_a)$ of a single atom at a frequency ω. This atom has a resonance frequency ω_a.

$$\tilde{\chi}_n(\omega, \omega_a) \propto \frac{1}{\underbrace{\Delta\omega_a}_{\text{linediwdth}} + 2i(\omega - \omega_a)} \quad (*)$$

Atoms around this frequency constitute a "spetral packet"

Reminder: Susceptibilities come from the steady state response of a medium to a field

$$D(\omega) = \epsilon_0 \underbrace{E(\omega)}_{\substack{\text{electric}\\ \text{field}}} + \underbrace{P(\omega)}_{\text{polarization}}$$

linear response:

$$\chi(\omega) \equiv \frac{P(\omega)}{\epsilon_0 E(\omega)}$$

In this framework, the atoms are approximated by harmonic oscillators, and in (*) we take the near-resonant case

Details: undergraduate electromagnetism course

See also Siegman, Lasers

Number of atoms in an inhomogeneously broadened line with resonant frequencies in a range $d\omega_a$ around ω_a

$$dN(\omega_a) = N\, g(\omega_a)\, d\omega_a$$

$g(\omega_a) \rightarrow$ linewidth (normally assumed to be a gaussian)

$$\int_{-\infty}^{\infty} g(\omega_a)\, d\omega_a = 1 \quad \text{(normalized)}$$

If saturation takes place near $\omega = \omega_a$, this means that a spectral wavepacket at a frequency ω_a will have its population reduced by the saturating factor

$$S(\omega_a) = \frac{1}{1 + F(\omega_a, \omega_1, I_1, I_{sat})}$$

with

$$F(\omega_a, \omega_1, I_1, I_{sat}) = \frac{I_1}{I_{sat}} \frac{1}{1 + \left[2(\omega_1 - \omega_a)/\Delta\omega_a\right]^2}$$

$N \rightarrow N\, S(\omega_a)$

Relatively easy to show: see, for instance, 2nd coursework for saturation in a 4-level system

I_1 = Strong saturating signal's intensity

I_{sat} = saturation intensity

Total susceptibility: $\tilde{\chi}(\omega) = N \int_{-\infty}^{\infty} g(\omega_a)\, S(\omega_a)\, \tilde{\chi}_n(\omega; \omega_a)\, d\omega_a$

sum over all resonance freq. ω_a

We are interested in the change in susceptibility when the saturating signal is turned on

$$\delta\tilde{\chi}(\omega) \equiv \underbrace{\tilde{\chi}_0(\omega)}_{\text{unsaturated susceptibility}} - \tilde{\chi}(\omega) = N \int_{-\infty}^{\infty} g(\omega_a)\left[1 - S(\omega_a)\right] \tilde{\chi}_n(\omega; \omega_a)\, d\omega_a$$

$$1 - S(\omega_a) = 1 - \frac{1}{1+F} = \frac{F}{F+1} = \frac{I}{I_{sat}} \left\{ \frac{\cancel{A}}{\cancel{A}\left[1 + \frac{I_1}{I_{sat}} + \left[1 + \left[\frac{2(\omega_1-\omega_a)}{\Delta\omega}\right]^2\right]\right.} \right\}$$

$$= \frac{I_1}{I_{sat}} \left[\frac{1}{1 + I_1/I_{sat} + \left(2(\omega-\omega_a)/\Delta\omega_a\right)^2} \right]$$

$$\delta \tilde{\chi}(\omega) \propto \int_{-\infty}^{\infty} g(\omega_1+\omega_a-\omega_1) \overbrace{\left[\frac{I_1/I_{sat}}{1 + I_1/I_{sat} + [2(\omega_1-\omega_a)/\Delta\omega_a]^2} \right]}^{\text{real lorentzian}} \times \underbrace{\left[\frac{1}{1 + 2i(\omega-\omega_a)/\Delta\omega_a} \right]}_{\text{complex lorentzian}} d\omega_a$$

for a strongly inhomogeneous line $g(\omega_1-\omega_a-\omega_1)$ varies much more slowly than the rest and can be pulled out of the integral.

The integral reads

$$\int_{-\infty}^{\infty} \frac{p}{1+p+x^2} \frac{1}{1+i(y-x)} dx = \frac{\pi p}{\sqrt{1+p}} \cdot \underbrace{\frac{1}{1+\sqrt{1+p}+iy}}_{\substack{\text{complex Lorenzian} \\ \text{depending on } \frac{\omega_a-\omega_1}{\Delta\omega_a}}}$$

$p = I_1/I_{sat}$

$x = 2(\omega_a-\omega_1)/\Delta\omega_a$

$y = 2(\omega-\omega_a)/\Delta\omega_a$

$dx = \frac{2}{\Delta\omega_a} d\omega_a$

By residues:

Poles at

$x = \pm i\sqrt{p+1}$ $\quad x = y - i$

$$\Rightarrow \delta\chi(\omega) \propto \frac{p}{\sqrt{1+p}} \frac{1}{1+\sqrt{1+p}+2i\frac{(\omega-\omega_1)}{\Delta\omega_a}} = \frac{p}{\sqrt{1+p}\,(1+\sqrt{1+p})} \cdot \frac{1}{1+\frac{2i(\omega-\omega_1)}{\Delta\omega_h}}$$

$$\Delta\omega_h = [1+\sqrt{1+p}]\,\Delta\omega_a$$

↳ Linewidth of the hole!

$I \ll I_{sat} \Rightarrow \Delta\omega_h = 2\Delta\omega_a$

$I \gg I_{sat} \Rightarrow \Delta\omega_h = \sqrt{I/I_{sat}}\,\Delta\omega_a$

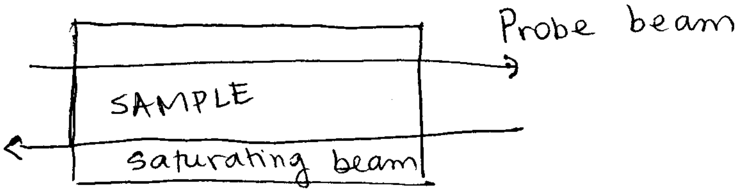
(*) Conclusions :

- The change in susceptibility produced by burning the hole is a complex Lorentzian line centered at $\omega = \omega_1$
- It is as if we had destroyed a certain number of atoms at $\omega = \omega_1$
- The linewidth of the hole broadens as the intensity increases, but is still much smaller than $g(\omega)$

For that reason, it is used in saturation absorption spectroscopy

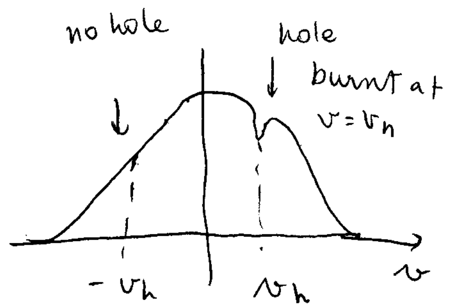
Saturation absorption spectroscopy

- Useful for finding homogeneous lineshapes hidden inside Doppler-broadened transitions
- set up: A strong saturating beam and a weak probe beam (normally both at freq. ω).



Let us consider a Gaussian (Maxwellian) distribution of axial velocities for a collection of atoms in a gas

The strong beam will burn a hole in the velocity distribution at $v = v_h = \frac{\omega_0 - \omega}{\omega_0} c$

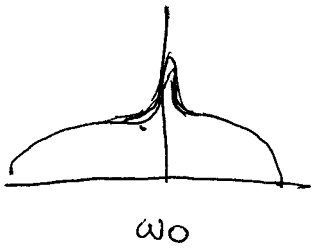


The probe wave will interact primarily with an oppositely traveling group of atoms so that

$$v = -v_h = -\frac{(\omega_0 - \omega)}{\omega_0} c$$

If, however, the frequency ω is tuned to the unshifted resonance frequency, then <u>both</u> lasers will interact with the <u>SAME</u> group of atoms (i.e., those with <u>ZERO</u> axial velocity)

→ Absorption will be reduced and there will be an increase in the intensity of the probe pulse at $\omega = \omega_0$

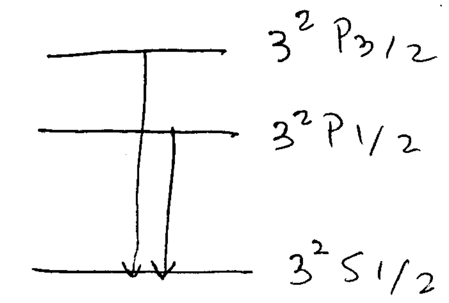


First observation: P. H. Lee and M. L. Skolnick, Appl. Phys. Lett. 10, 303 (1967) (He-Ne laser)

Saturation absorption spectroscopy using dye lasers:

T. Hänsch, I S Shahin and A. L. Schawlow, Phys. Rev. Lett. 27, 707 (1971)

Sodium lines



Lamb shift : associated to the fluctuation of the zero-point energy
↳ greatest for states in which $|\psi(0)|^2 \neq 0$
$n\,^2S_{1/2}$ states. This is normally obscured by the Doppler broadening

copy page 456, Corney

Hänsch et al, PRL 34, 307 (1975)

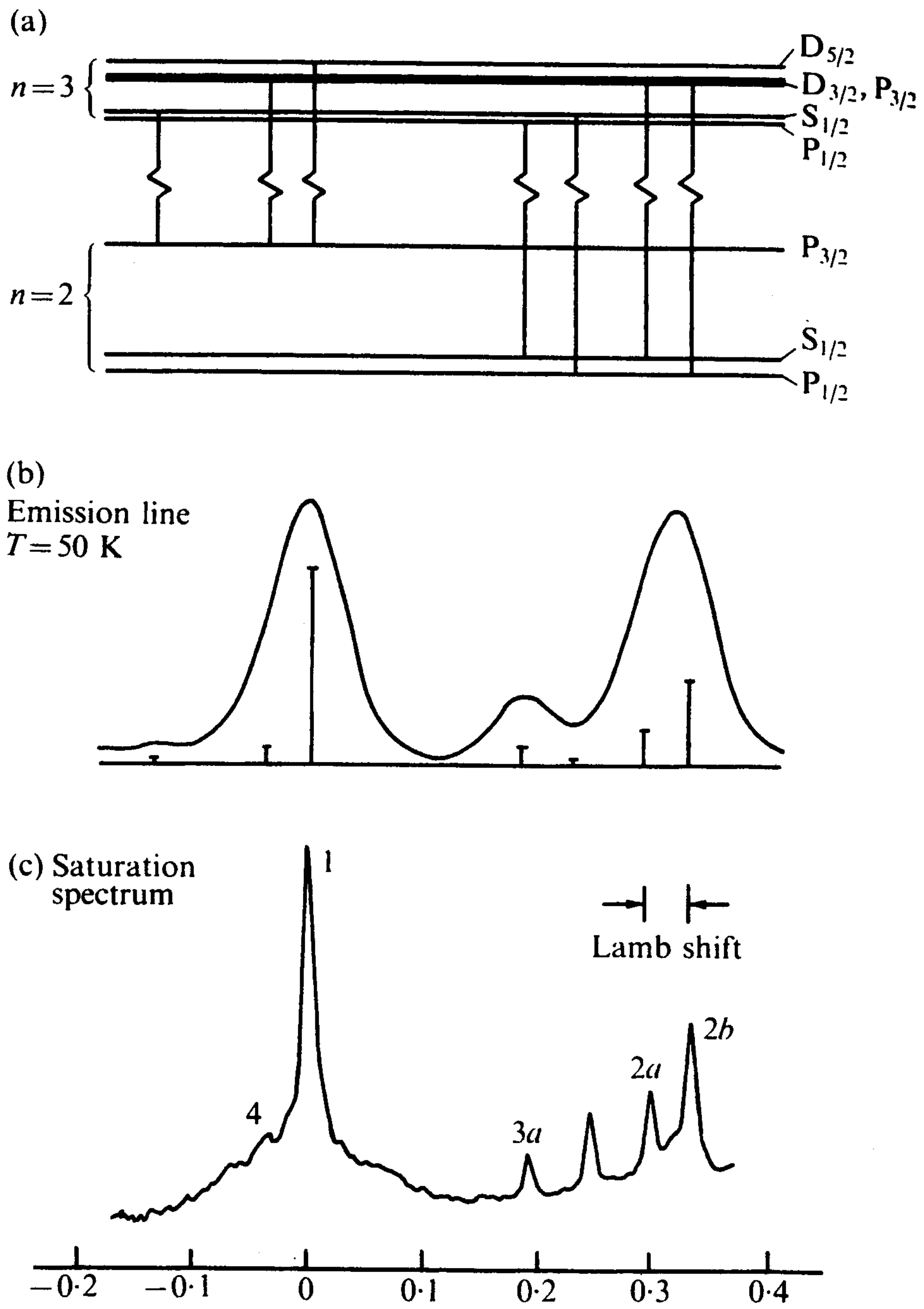


Fig.14.7. Fine structure in hydrogenic systems - the Balmer α line of deuterium. (a) Energy levels and allowed fine-structure transitions. (b) Profile of emission line from cooled deuterium gas discharge and fine structure components with theoretical relative transition probabilities. (c) Saturation spectrum of D_α line showing optically resolved Lamb shift. (After Hänsch (1975).)

5. (b) - Doppler - free two photon spectroscopy

(*) key idea: one employs a two-photon transition to eliminate the doppler broadening

Let us consider an atom irradiated by two laser beams of
- Frequencies : ω, ω'
- Polarization vectors: $\hat{\varepsilon}, \hat{\varepsilon}'$

$$\vec{A}(\vec{r},t) = A_0 \hat{\mathcal{E}} \cos(\omega t - \vec{k}\cdot\vec{r}) + A_0' \hat{\mathcal{E}}' \cos(\omega t - \vec{k}'\cdot\vec{r})$$

where $\vec{k} = \frac{\omega}{c}\hat{k}$; $\vec{k}' = \left(\frac{\omega'}{c}\right)\hat{k}'$

This atom, initially in a state a, absorbs two photons and is excited to the state b.
Let us initially assume that the atom is stationary

(+) Transition rate (two-photon absorption)
(obtained from 2nd-order perturbation theory)

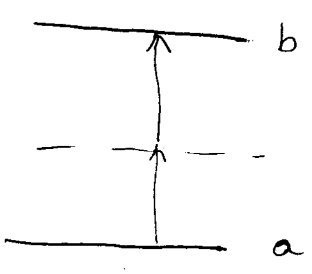
$$W_{ba} = \frac{4\pi^2}{c^2 \varepsilon_0^2} I I' \left| \sum_n \left\{ \frac{(\hat{\varepsilon}'\cdot\vec{D}_{bn})(\hat{\varepsilon}\cdot\vec{D}_{na})}{\omega_{na}-\omega} + \frac{(\hat{\varepsilon}\cdot\vec{D}_{bn})(\hat{\varepsilon}'\cdot\vec{D}_{na})}{\omega_{na}-\omega'} \right\} \right|^2 \times \delta(\omega_{ba}-\omega-\omega') \quad (*)$$

where

$I, I' \equiv$ intensities of the 2 laser beams

$\omega_{na} = \frac{(E_n - E_a)}{\hbar}$, $\omega_{ba} = \frac{(E_b - E_a)}{\hbar}$,

$n \equiv$ intermediate levels $\left(\begin{matrix} n \neq b \\ n \neq a \end{matrix}\right)$



(*) Assumptions all intermediate states are non-resonant
intermediate st. are virtual states

(*) Consequences :

- The atom will decay from b by a single-photon transition

Excited state b: width $= \Gamma_b$

lifetime $\tau_b = \frac{\hbar}{\Gamma_b}$

$$\omega_{ba} \to \omega_{ba} - \frac{i\Gamma_b}{2\hbar}$$

$$\delta(\omega_{ba} - \omega' - \omega) \to \frac{1}{\pi} \frac{\Gamma_b/(2\hbar)}{\pi(\omega_{ba} - \omega - \omega')^2 + (\Gamma_b/2\hbar)^2}$$

- I, I' and the quantities in { } in (*) will vary slowly

$\Rightarrow$ The line profile for two photon absorption will be given by

$$g(\omega, \omega') = \frac{\Gamma_b/(2\hbar)}{(\omega_{ba} - \omega - \omega')^2 + (\Gamma_b/2\hbar)^2}$$

Unrealistic: atoms are moving with velocity v

$\Rightarrow$ $\vec{r} = \vec{v}t$

and
$$A(\vec{r}, t) = A_0 \hat{\varepsilon} \cos\left[\omega\left(1 - \frac{\vec{v}\cdot\hat{k}}{c}\right)t\right] + A_0' \hat{\varepsilon}' \cos\left[\omega'\left(1 - \frac{\vec{v}\cdot\hat{k}'}{c}\right)t\right]$$

By inspection we see that

$$\omega \to \omega\left(1 - \vec{v}\cdot\frac{\hat{k}}{c}\right)$$
$$\omega' \to \omega'\left(1 - \vec{v}\cdot\frac{\hat{k}}{c}\right)$$

$$g(\omega, \omega') = \frac{\Gamma/(2\hbar)}{\left[\omega_{ba} - \omega(1 - \vec{v}\cdot\hat{k}/c) - \omega'(1 - \vec{v}\cdot\hat{k}'/c)\right]^2 + \Gamma(2\hbar)^2}$$

In general, one must integrate over the velocity distribution
$\Rightarrow$ Doppler broadening.

Let us now assume that the two beams propagate in opposite directions and have the same angular frequency

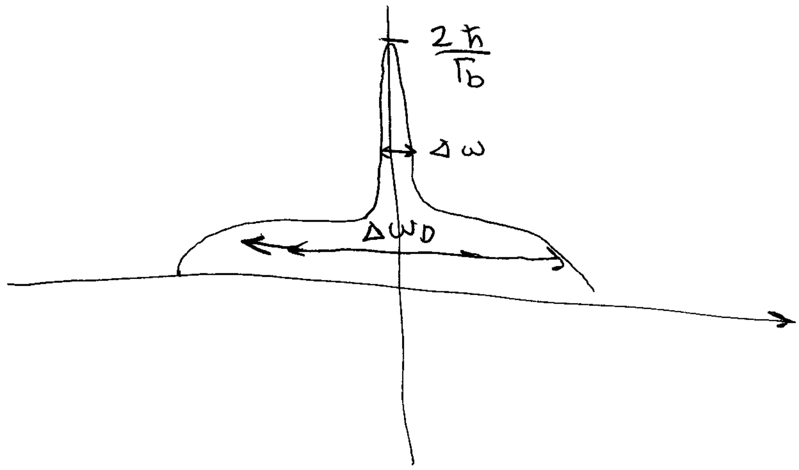
$$\Rightarrow \vec{k} = -\vec{k}' , \quad \omega = \omega'$$

$$\Rightarrow \boxed{g(\omega, \omega) = \frac{\Gamma_b/(2\hbar)}{(\omega_{ba} - 2\omega)^2 + [\Gamma_b/(2\hbar)]^2}}$$

- The Doppler broadening has been eliminated!
  - Maximum: $\omega = \frac{\omega_{ba}}{2}$
  - width: $\frac{\Gamma_b}{\hbar} = \Delta\omega$

$\rightarrow$ This peak is superimposed on a much broader Doppler distribution arising from the absorption of 2 photons from either beam

$$\Delta\omega^D = \frac{\omega_{ba}}{c} \left(\frac{2 k_b T \ln 2}{M} \right)^{1/2}$$

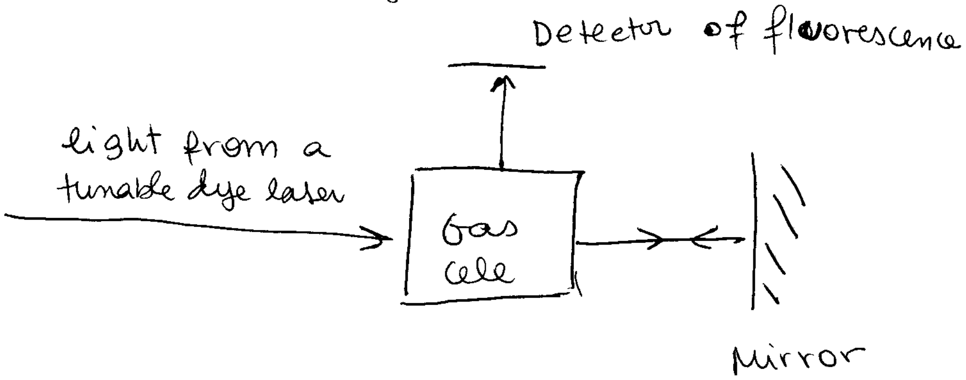


In experiments

① The frequency of the laser is swept through the region centered on $\omega = \omega_{ba}$ (dye laser)

② The ~~frequency of the laser~~ counter-propagating beam of the same frequency

Obtained by using a mirror



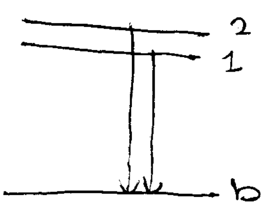
Examples

- Measurement of the $1s \rightarrow 2s$ excitation - T. Hänsch, S. A. Lee, C. Wallenstein and C. E. Wieman, Phys. Rev. Lett. 34, 307 (1975)
  - Doppler - free two-photon $1s \rightarrow 2s$ line
  - Hyperfine Doublet for ~~F=1 and~~ $F = 1 \rightarrow 1$, $F = 0 \rightarrow 0$ transitions
  - Lamb shift of the $1s$ state

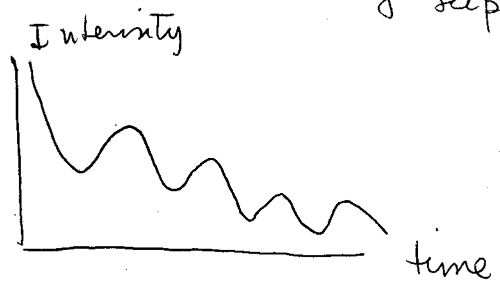
⚠ T. Hänsch ⇒ Nobel prize, 2005

5. (c) - Quantum beat spectroscopy

(*) Assumptions :



- Atom with a set of states closely spaced in energy : if these states can be excited coherently the time dependence of the radiation will be that of a decreasing exponential modulated by superimposed oscillations ⇒ "Quantum beats"



Time-dep. wavefunction

$$|\Psi(t)\rangle = c_1 |1\rangle \exp[-iE_1 t/\hbar - t/(2\tau)] + c_2 |2\rangle \exp[-iE_2 t/\hbar - t/(2\tau)]$$

⊛ Intensity of the decay radiation (dipole approximation)

$$I(t) = A \, |\langle b | \hat{\varepsilon} \cdot \vec{D} | \psi(t) \rangle|^2$$

$$= A \Big\{ |C_1|^2 \, |\hat{\varepsilon} \cdot \underbrace{\langle b | \vec{D} | 1 \rangle}_{\vec{D}_{b1}}|^2 + |C_2|^2 \, |\hat{\varepsilon} \cdot \underbrace{\langle b | \vec{D} | 2 \rangle}_{\vec{D}_{b2}}|^2 +$$

$$+ 2 |C_1| |C_2| \, |\hat{\varepsilon} \cdot \vec{D}_{b1}| \, |\hat{\varepsilon} \cdot \vec{D}_{b2}| \cos\left[\frac{(E_2 - E_1) t}{\hbar} + \varphi \right] \cdot$$

$$\cdot \exp[-t/\tau]$$

⇒ from the oscillations one can measure the energy difference $E_2 - E_1$

– **Example** : Modulation of the decay

$5\,^3P_1 (M_J = 0) \rightarrow 5\,^1S_0$

$5\,^3P_1 (M_J = 1) \rightarrow 5\,^1S_0$ of Cd

(Zeeman shift induced by a magnetic field)

$\omega_{21} = (E_2 - E_1)/\hbar = \frac{g \mu_0 B}{\hbar}$ ⇒ ω_{21} enables the determination of the Landé factor

– J. N. Dodd, W. J. Sandle and D. Zissermann, Proc. Phys. Soc. (London) 92, 497 (1967).