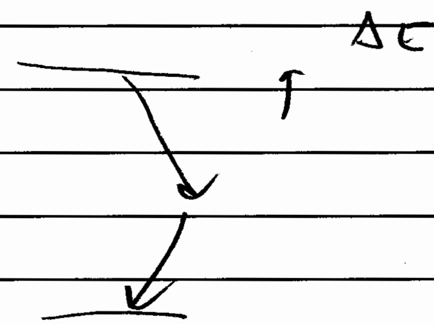
What is the virtual state?

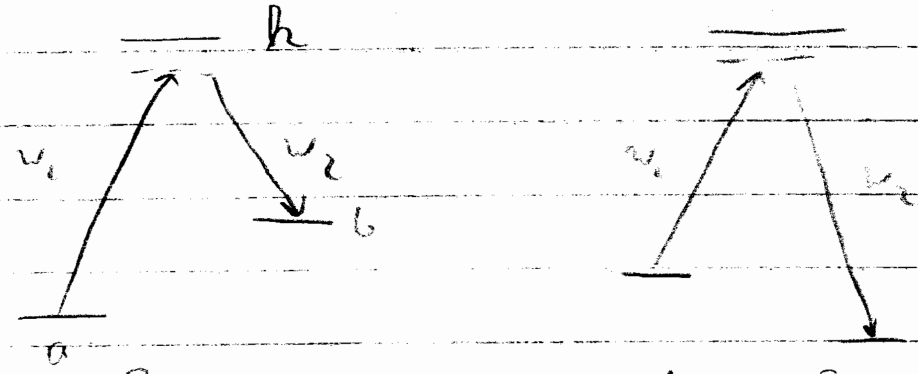
- It is what the perturbation sum says
- It is a component of the dressed atom wave function
- It can be a linear superposition of several states (but <u>all</u> at the frequency of the drive
- Lifetime: <u>gamma</u> two-photon spont. e




intermediate state high up with ΔE

2 photons are correlated to within $\hbar/\Delta E$

## Raman process




Stokes          Anti-Stokes

$\omega_1 = \omega_2 + \omega_{ba}$

correspond to 2-ph. absorption

since one photon is emitted ($\omega_2$) we have to keep the $e^{i\omega_2 t}$ term

(which, for absorption, is the counterrotating term)

Note $\frac{1}{i} E_1 \left(e^{i\omega_1 t} + e^{-i\omega_1 t}\right)$ } total of four terms

↳ $(\ )\left(a_1 + a_1^+\right)$ } two co-rotating, two counter-rotating

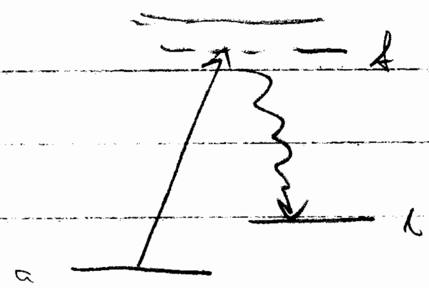
everything analogous to 2-photon absorption, but $-\omega_2 \to +\omega_2$

detuning $\Delta\omega = \omega_0 - \omega_1 + \omega_2$

$$\Gamma_{ab}^{Raman} = \frac{\pi}{2} \omega_{R2}^2 \, \delta(\Delta\omega) \qquad \omega_{R2} = \frac{\omega_R^{(1)} \omega_R^{(2)}}{2\Delta}$$

## Spontaneous Raman process

Before lasers came up which could stimulate emission process.
Spont. Raman scattering was the only relevant two-photon process.




pop. of virtual state (either perturbation or dressed atom)

$$\frac{w_R^2}{4\Delta^2}$$

(Note: can be obtained as a transition rate $\frac{w_R^2}{\Delta}$ × the dwell time $\tau \sim 1/\Delta$)

A coeff. for em.: $A_{fb}$

$$\Gamma_{ab}^{\text{spont Ra}} = \frac{w_R^2}{4\Delta^2} A_{fb}$$

Note: even if $a = b$, $b$ differs from $a$ by the photon recoil ⇒ Res. fluorescence and Rayleigh scattering are Raman processes!

# Two-photon spectroscopy

## Doppler effect in two-photon spectroscopy

two beams $\omega_1, \vec{k}_1$ $\quad \omega_2, \vec{k}_2$

atom moves with $v$

$\omega_i' = \omega_i - \vec{k}_i \cdot \vec{v}$

$$\Gamma_{ab} = \frac{\pi}{2} \omega_{R2}^2 \, f(\Delta\omega)$$

$$f(\omega_1 + \omega_2) = \frac{2}{\pi} \frac{(\gamma/2)}{(\gamma/2)^2 + (\omega_0 - \omega_1' \pm \omega_2')^2}$$ Lorentzian

⟶ $-$ 2ph Abs, $+$ Raman

$$= \frac{2}{\pi} \frac{(\gamma/2)}{(\gamma/2)^2 + \left[\omega_0 - (\omega_1 \pm \omega_2) + (\vec{k}_1 \pm \vec{k}_2)\cdot\vec{v}\right]^2}$$

2-Ph Abs or Raman transition is like single-photon trans. with $\omega = \omega_1 \pm \omega_2$

$\vec{k} = \vec{k}_1 \pm \vec{k}_2$

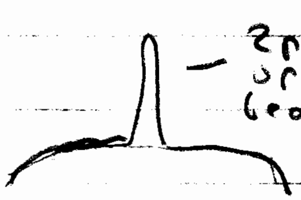
Doppler shift $\Delta\omega_D = (\vec{k}_1 \pm \vec{k}_2)\cdot\vec{v}$

minimized for
$\hat{k}_1 = -\hat{k}_2$ (abs)
$\hat{k}_1 = \hat{k}_2$ (Raman)

If $\omega_1 = \omega_2$ ($|k_1| = |k_2|$)

$\Delta\omega_D = 0$ Doppler free two-photon spectroscopy!

realized in standing wave for which $\vec{k}_1 = -\vec{k}_2$



Residual Doppler broadening only in second order

$$\delta\omega_{D2} = -\frac{1}{2}\frac{v^2}{c^2}\,\omega$$

$$\frac{\delta\omega_{D2}}{\omega} = -\frac{1}{2}\frac{Mv^2}{Mc^2} \simeq \frac{-kT}{Mc^2}$$ Room temp and hyd: $2\times10^{-11}$

For spectroscopy at $10^{-13}$: Cooling to millikelvin essential (but sufficient)

Famous example: 1s → 2s transition

Lifetime 1/7 sec ⇒ 1 Hz natural linewidth

narrowest linewidth: $\lesssim$ few kHz

Hänsch — 2nd order Doppler shift

Kleppner / Greytak — $\lesssim$ 350 Hz

↑ Most important systematics