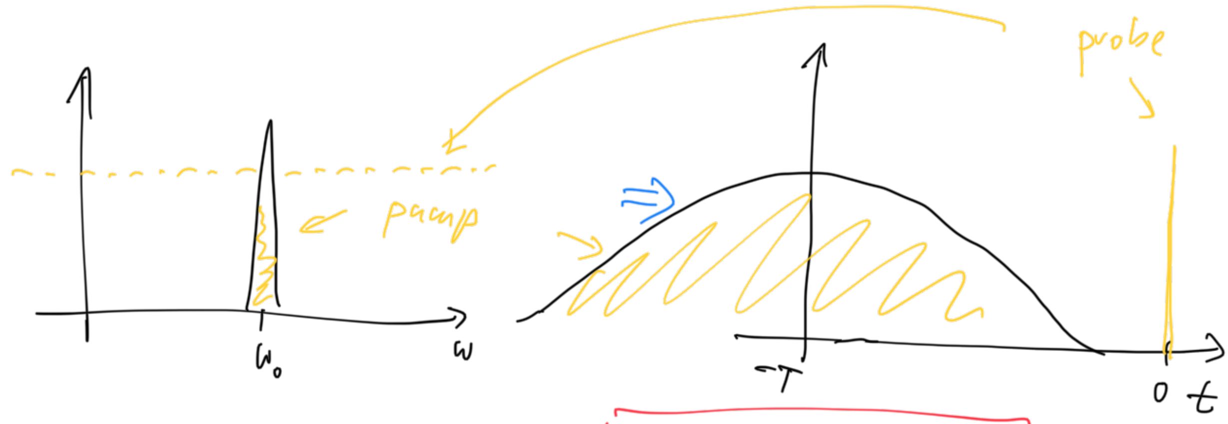
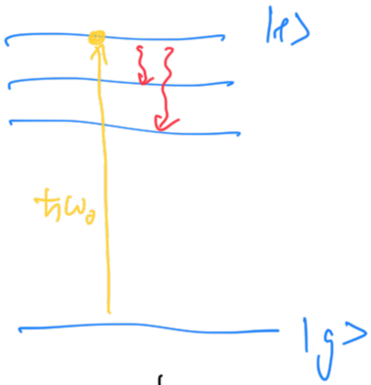
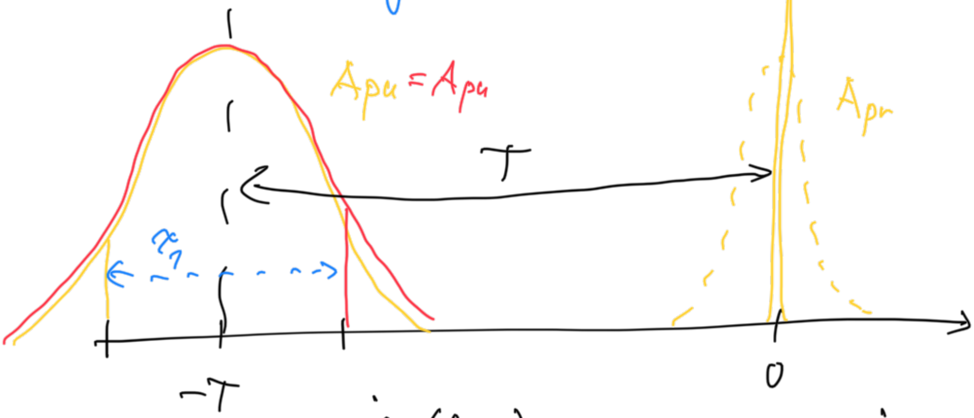
Pump-probe with finite pulses



$$\boxed{\Delta\omega \Delta T \approx \text{const.}}$$





$$A_{pu}(t+T)\, e^{-i\omega(t+T)} \qquad A_{pr}(t)\, e^{-i\omega t}$$

- Pump pulse occurs 2× in the perturbation theory
- Pump precedes probe

$$\underline{P^{(3)}(t, T, \tau = 0)} \approx \int_0^\infty d\tau_3 \int_0^\infty d\tau_2 \int_0^\infty d\tau_1 \, S^{(3)}(\tau_3, \tau_2, \tau_1) E(t-\tau_3) E(t-\tau_3-\tau_2) \times E(t-\tau_3-\tau_2-\tau_1)$$

- the response is a sum of Liouville pathways
- we look into the direction $-\vec{k}_1+\vec{k}_2+\vec{k}_3 = -\vec{k}_{pu}+\vec{k}_{pu}+\vec{k}_{pr}=\vec{k}_{pr}$
- in this direction we consider interaction order

1 – 2 – 3 ← rephasing pathways

2 – 1 – 3 ← non-rephasing pathways

$-\vec{k}_1+\vec{k}_2+\vec{k}_1$

$$P^{(3)}(t,T,0) \approx (i)^3 \underline{e^{-i\omega(t-\overset{=0}{\tau})}} \int_0^\infty d\tau_3 \int_0^\infty d\tau_2 \int_0^\infty d\tau_1 \; \overset{\approx e^{-i\omega_{eg}(\tau_3-\tau_1)}}{R_{(reph)}(\tau_3,\tau_2,\tau_1)}$$

1–2–3
$$\times e^{+i\omega(\tau_3-\tau_1)} A_3(t-\tau_3)\, A_2(\underline{t+T}-\underline{\tau_3-\tau_2})\, A_1^*(\underline{t+T}-\tau_3-\underline{\tau_2-\tau_1})$$

pr   pu $\tau=0$

$$+ (i)^3 e^{-i\omega(t-\overset{=0}{\tau})} \int_0^\infty d\tau_3 \int_0^\infty d\tau_2 \int_0^\infty d\tau_1 \; \overset{\approx e^{-i\omega_{eg}(\tau_3+\tau_1)}}{R_{(non\text{-}reph)}(\tau_3,\tau_2,\tau_1)}$$

2–1–3
$$\times e^{+i\omega(\tau_3+\tau_1)} A_3(\underline{t-\tau_3})\, A_1^*(\underline{t+T-\tau_3-\tau_2})\, A_2(\underline{t+T-\tau_3-\tau_2-\tau_1})$$

$$\boxed{A_3(t) = A_{pr}(t) \approx A_{pr}\,\delta(t)}$$

$$\boxed{R(t,\tau_2,\tau_1) = \mathcal{U}(t)\,\mathcal{U}(\tau_2)\,\mathcal{U}(\tau_1)}$$

$$P^{(3)}(t,T) \approx (i)^3 \cancel{e^{-i\omega t}} A_{pr} \int_0^\infty d\tau_2 \int_0^\infty d\tau_1 \; R_{(reph)}(t,\tau_2,\tau_1)\, e^{+i\omega(\cancel{t}-\tau_1)}$$

$$\times A_2(\cancel{t}+T-\cancel{t}-\tau_2)\, A_1^*(\cancel{t}+T-\cancel{t}-\tau_2-\tau_1)$$

$$+ \ \dots$$

$$= (i)^3 A_{pr}\, \mathcal{U}(t) \int_0^\infty d\tau_2\, \mathcal{U}(\tau_2)\, A_2(T-\tau_2) \int_0^\infty d\tau_1\, \underbrace{\mathcal{U}(\tau_1)\, e^{-i\omega\tau_1}}_{f} \times \underbrace{A_1^*(T-\tau_2-\tau_1)}_{g(\dots)}$$

$$x = T-\tau_2$$

$$\int_0^\infty d\tau_1\, f(\tau_1)\, g(x-\tau_1)$$

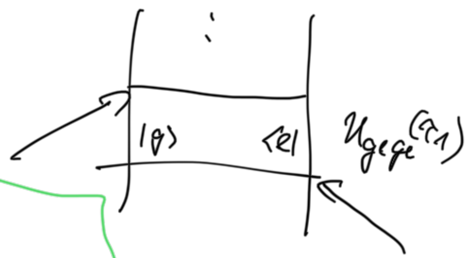
Convolution $\quad [f*g](t) = \int_0^\infty d\tau\, f(\tau)\, g(t-\tau)$

$$\boxed{\int_{-\infty}^{\infty} dx\, e^{i\omega x} [f*g](x) = f(\omega)\, g(\omega)}$$

$$f(\omega) = \int_{-\infty}^{\infty} d\tau\, f(\tau)\, e^{i\omega\tau}$$

$$\boxed{[f*g](x) = \int_{-\infty}^{\infty} d\omega\, e^{-i\omega x} f(\omega)\, g(\omega)}$$!

Rephasing diagrams



Complex lineshape

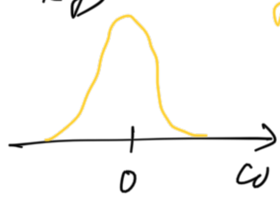
$$\boxed{\mathcal{U}_{gege}(\tau_1)\, e^{-i\omega\tau_1} = \tilde{\mathcal{U}}_{gege}(\tau_1)\, e^{i(\omega_{eg}-\omega)\tau_1}}$$

$$\int_{-\infty}^{\infty} d\omega\, \mathcal{U}_{egeg}(\tau)\, e^{i\omega\tau} = \int_{-\infty}^{\infty} d\omega\, \tilde{\mathcal{U}}_{egeg}(\tau)\, e^{-i\omega_{eg}\tau + i\omega\tau}$$

$$= G_{eg}(\omega-\omega_{eg})$$

centered around ω_{eg}

$$\boxed{G_{eg}(\omega) = \int_{-\infty}^{\infty} d\omega\, \tilde{\mathcal{U}}_{egeg}(\tau)\, e^{i\omega\tau} =}$$



We have

$$\mathcal{U}_{gege}(\tau_1)\, e^{-i\omega\tau_1} \xrightarrow{iFT} \int_{-\infty}^{\infty} d\tau_1 \left[\hat{\tilde{\mathcal{U}}}_{gege}(\tau_1)\, e^{i\omega_{eg}\tau_1 - i\omega\tau_1}\right] e^{-i\omega'\tau_1}$$

$$= \int_{-\infty}^{\infty} d\tau_1\, \hat{\tilde{\mathcal{U}}}_{gege}(\tau_1)\, e^{i(\omega_{eg}-\omega-\omega')\tau_1}$$

$$= \int_{-\infty}^{\infty} d\tau_1 \left[\hat{\tilde{\mathcal{U}}}_{egeg}(\tau_1)\, e^{i(\omega'+\omega-\omega_{eg})\tau_1}\right]^*$$

$$\boxed{= G^*_{eg}(\omega'+\omega-\omega_{eg})} =$$

$$= G^*_{eg}(\omega'-\Delta) \qquad \boxed{\Delta = \omega_{eg}-\omega}$$

$$A^*_1(T-\tau_2-\tau_1) \xrightarrow{iFT} \int_{-\infty}^{\infty} d\tau_1\, A^*_1(\tau_1)\, e^{-i\omega'\tau_1} = \int_{-\infty}^{\infty} d\omega' \left[A_1(\tau)\, e^{i\omega'\tau}\right]^*$$

$$\boxed{= A^*_1(\omega')}$$

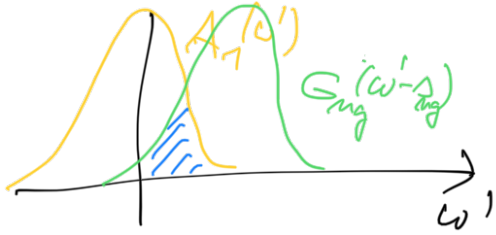
$$\mathcal{P}^{(3)}(t,T) \approx (i)^3 A_{pr}\, \mathcal{U}(t) \int_0^{\infty} d\tau_2\, \mathcal{U}(\tau_2)\, A_2(T-\tau_2)$$

$$\times \int_{-\infty}^{\infty} d\omega'\, e^{-i\omega'(T-\tau_2)}\, G^*_{mg}(\omega'-\underbrace{(\omega_{eg}-\omega)}_{\Delta_{eg}})\, A^*_1(\omega')$$

... detuning of pulse from transition

$+ \ldots$

$$G^*_{mg}(\omega'-\Delta_{eg})\, A^*_1(\omega')$$



Non-rephasing part

the only difference is $e^{i\omega\tau_1}$

pulse A_2

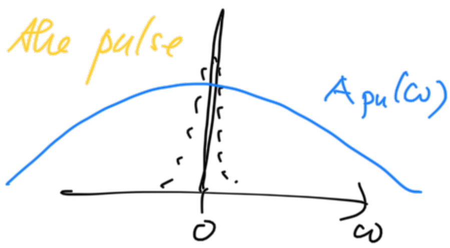
$$P^{(3)}(t,T,\tau=0)\Big|_{non-reph} \approx (i)^3 A_{pr}\, u(t) \int_0^\infty d\tau_2\, u(\tau_2)\, A_1^*(t-\tau_2)$$

$$\times \underline{\int_0^\infty d\tau_1\, u_{eg}(\tau_1)\, e^{i\omega\tau_1} A_2(T-\tau_2-\tau_1)}$$

$$= (i)^3 A_{pr}\, u(t) \int_0^\infty d\tau_2\, u(\tau_2)\, A_1^*(T-\tau_2) \int_{-\infty}^{\infty} d\omega'\, e^{i\omega'(T-\tau_2)}\, \underline{G_{eg}(\omega'+\Delta_{eg})}\, \underline{A_2(\omega')}$$

Case: line-shape narrower than the pulse

$$G_{eg}(\omega) \approx \delta(\omega)$$



$\omega' = \Delta_{eg}$

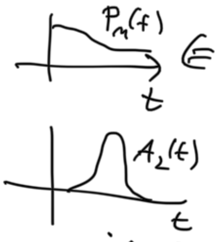
$$\int_{-\infty}^{\infty} d\omega'\, e^{-i\omega'(T-\tau_2)}\, \delta(\omega'+\omega-\omega_{eg})\, A_1^*(\omega') =$$

$$= e^{-i\underbrace{(\omega_{eg}-\omega)}_{\Delta_{eg}}(T-\tau_2)}\, A_1^*(\Delta_{eg})$$

$$P^{(3)}(t,T) \simeq (i)^3 A_{pr}\, \mathcal{U}(t) \int_0^\infty d\tau_2\, \mathcal{U}(\tau_2)\, A_2(T-\tau_2)\, A_1^*(\Delta_{1g})\, e^{-i\Delta_{eg}(T-\tau_2)}$$

$$+ \dots$$

$\mathcal{U}(\tau_2) \approx \text{const.}$ $= \mathcal{U}(T)$



$\in \mathcal{U}(t)$

$$= (i)^3 A_{pr}\, \mathcal{U}(t)\, \mathcal{U}(\underset{T}{\cancel{\tau_2}})\, A_1^*(\Delta_{1g}) \int_0^\infty d\tau_2\, A_2(T-\tau_2)\, e^{-i\Delta_{eg}(T-\tau_2)}$$

$\tau' = T - \tau_2$

$d\tau' = -d\tau_2$

$\tau'(\infty) = -\infty$

$\tau'(0) = T$

$$+ \dots$$

$$= (i)^3 A_{pr}\, \mathcal{U}(t)\, \mathcal{U}(\underset{T}{\cancel{\tau_2}})\, A_1^*(\Delta_{1g}) \underbrace{\int_{-\infty}^{T} d\tau'\, A_2(\tau')\, e^{-i\Delta_{eg}\tau'}}_{A_2(\Delta_{eg})}$$

$$+ \dots$$

$$=$$

$$P^{(3)}(t,T) \approx (i)^3 A_{pr}\, \mathcal{U}(t)\, \mathcal{U}(\underset{T}{\cancel{\tau_2}})\, A_1^*(\Delta_{1g}) \int_{-\infty}^{T} d\tau'\, A_2(\tau')\, e^{-i\Delta_{eg}\tau'}$$

$$+ (i)^3 A_{pr}\, \mathcal{U}(t)\, \mathcal{U}(\underset{T}{\cancel{\tau_2}})\, A_2(\Delta_{1g}) \int_{-\infty}^{T} d\tau'\, A_1^*(\tau')\, e^{i\Delta_{eg}\tau'}$$

$\boxed{A_1 \text{ and } A_2 \text{ are real and equal to } A}$

$$\approx (i)^3 A_{pr}\, \mathcal{U}(t)\, \mathcal{U}(\tau_2)\, A(\Delta_{eg}) \left[\int_{-\infty}^{T} d\tau' A(\tau') e^{-i\Delta_{eg}\tau'} + \int_{-\infty}^{T} d\tau' A(\tau') e^{i\Delta_{eg}\tau'} \right]$$

Sub.

$\tau'' = -\tau'$

$d\tau'' = -d\tau'$

$\tau''(-\infty) = \infty$

$\tau''(T) = -T$

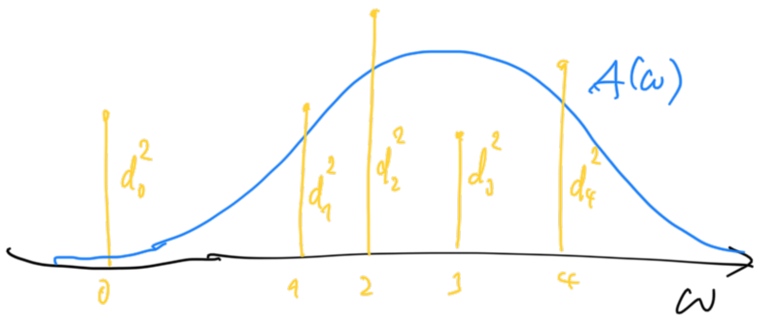
$$\int_{\infty}^{-T} (-d\tau'') \rightarrow \int_{-T}^{\infty} d\tau'' \underbrace{A(-\tau'')}_{= A(\tau'')} e^{i\Delta_{eg}\tau''}$$

symmetric pulse

$$\boxed{P^{(3)}(t,T) \approx (i)^3 A_{pr}\, \mathcal{U}(t)\, \mathcal{U}(\tau_2)\, A(\Delta_{eg}) \left[\int_{-\infty}^{\infty} d\tau' A(\tau') e^{i\Delta_{eg}\tau'} + \int_{-T}^{T} d\tau' A(\tau') e^{i\Delta_{eg}\tau'} \right]}$$

$T >$

$$\boxed{\approx (i)^3 A_{pr}\, \underline{\mathcal{U}(t)\, \mathcal{U}(\tau_2)}\, 2 |A_{pu}(\Delta_{eg})|^2}$$



Pump-probe spectrum can be devided by $|A_{pu}(\omega)|^2 = |E_{pu}(\omega)|^2$ to correct for pulse width (pulse spectrum)!

Erratum

$$\tau_2 \longrightarrow T$$