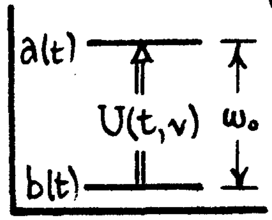
1992 $\Phi$507 Problems Assigned: 4/6. Due: 4/13/92. file HB11 **(11)**




(37) [20 pts]. Ref. class notes on tD Pert$^{\underline{b}}$n Theory, pp. tD 11-12. A two-level QM system (energy gap $\hbar\omega_0$) is subjected to a "chirped" coupling pulse $U(t,\nu) = \mathcal{E}(t)\, e^{-i[\nu - \theta(t)]t}$. The envelope $\mathcal{E}(t)$ has finite duration $\sim T$, and the main frequency $\nu \sim \omega_0$ drives transitions $b \to a$ as usual. What's new is that the "chirp" fcn $\theta(t)$ can <u>modulate</u> $\nu$ during the pulse.

(A) Find the spectral fcn $\delta$ corresponding to the rf carrier $e^{-i[\nu-\theta(t)]t}$ in the case where the chirp is: $\theta(t) = \Delta\nu \cdot \frac{t}{\tau}$, w/ $\Delta\nu$ (bandwidth) & $\tau$ (risetime) = cnsts. Show: $\underline{\delta(\omega) = (\alpha/\sqrt{\pi})\, e^{i\pi/4}\, e^{-i\alpha^2\omega^2}}$, and find $\alpha$ in terms of $\Delta\nu$ & $\tau$. Show that $\delta$ becomes a Dirac delta fcn as $\alpha \to \infty$. What is the significance of this limit?

(B) If the envelope fcn is $\mathcal{E}(t) = \mathcal{E}_0\, e^{-(t/T)^2}$, find the transition amplitude $a(\Omega)$ for the chirp of part (A). $\Omega = \omega_0 - \nu$ is the detuning frequency.

(C) Analyse the transition lineshape, i.e. $|a(\Omega)|^2$ vs. $\Omega$. Under what conditions on $\Delta\nu$, $\tau$ & $T$ does the envelope dominate $|a(\Omega)|^2$? When does the chirp dominate?

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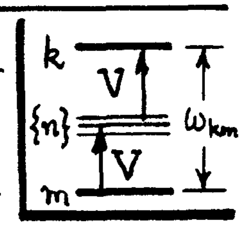


(38) [20 pts.]. A pulsed harmonic perturbation $V(x,t) = 2\hbar\Omega(x)\cos\omega t$, over $0 \leq t \leq T$, drives QM transitions $m \to k$. In class [class notes p. tD6, Eq.(18)], we found the 1st order transition amplitude $a_k^{(1)}(t)$, which describes direct single-photon $m \to k$ processes. Here we analyse the 2nd order amplitude $a_k^{(2)}(t)$, describing two-photon processes: $m \to \{n\}$, $\{n\} \to k$, through a set of intermediate states $\{n\}$. To fix ideas, let the transition be <u>absorptive</u>, and let the driving frequency $\omega \sim \omega_0$, w/ $\omega_0 = \frac{1}{2}\omega_{km} > 0$ = <u>half</u> the resonant freq. Define the detuning as $\Delta\omega = \omega - \omega_0$.

(A) Calculate the 2nd order amplitude $a_k^{(2)}(t)$ for the pulsed harmonic pert$^{\underline{b}}$n $V(x,t)$.

(B) Denote $\delta_{nm} = \omega - \omega_{nm}$. Show that the <u>resonant</u> parts of $a_k^{(2)}(t)$ contribute:

$$\boxed{a_k^{(2)}(t) \simeq \sum_n \frac{\Omega_{kn}\Omega_{nm}}{\delta_{nm}} \left[ \frac{1 - e^{-i(2\Delta\omega - \delta_{nm})t}}{2\Delta\omega - \delta_{nm}} - \frac{1 - e^{-i(2\Delta\omega)t}}{2\Delta\omega} \right]}, \text{ for } m \to \{n\} \to k \text{ @ } \omega \simeq \frac{\omega_{km}}{2}.$$

(C) In $a_k^{(2)}(t)$ of part (B), we can have $\Delta\omega \to 0$ (by tuning) and $\delta_{nm} \to 0$ (by "accident"). Find the limiting forms of $a_k^{(2)}(t)$ for the following 3 cases: (I) $\delta_{nm} \to 0$, for some $n$, and $\Delta\omega \to 0$; (II) $\delta_{nm} \to 0$, for some $n$, but $\Delta\omega \neq 0$; (III) $\delta_{nm} \neq 0$, for any $n$, while $\Delta\omega \to 0$. Show that $a_k^{(2)}(t)$ is always finite, but its behavior depends critically on the $\delta_{nm}$.

(37) [20 pts]. Find transition lineshape for a "chirped" coupling pulse.

Ref. pp. tD 11-12 of class notes on Time-Dependent Perturbation Theory.

(A) **1)** Let the coupling pulse be : $U(t,\nu) = \mathcal{E}(t)\{e^{-i[\nu-\theta(t)]t}\}$. The exponential is identified with the spectral fcn $\delta$ by $\{\ \} = \int_{-\infty}^{\infty} \delta(\omega-\nu) e^{-i\omega t} d\omega$, so we have

$$\rightarrow \delta(\omega-\nu) = \frac{1}{2\pi}\int_{-\infty}^{\infty}\left(e^{-i[\nu-\theta(t)]t}\right)e^{i\omega t}dt, \text{ or } 2\pi\delta(\kappa) = \int_{-\infty}^{\infty} e^{i[\kappa+\theta(t)]t}dt. \quad (1)$$

(w/ $\kappa = \omega - \nu$) by Fourier inversion. If the "chirp" fcn $\underline{\underline{\theta(t) = \Delta\nu\cdot\frac{t}{\tau}}}$, then...

$$\rightarrow 2\pi\delta(\kappa) = \int_{-\infty}^{\infty} e^{i[(\Delta\nu/\tau)t^2+\kappa t]}dt = \left(\int_0^{\infty} + \int_{-\infty}^{0}\right)dt\, e^{i[at^2+2bt]} \quad \int \begin{matrix} a = (\Delta\nu/\tau) \\ 2b = \kappa \end{matrix}$$

$$= \int_0^{\infty} dt\left\{e^{i(at^2+2bt)} + e^{i(at^2-2bt)}\right\}$$

integrals tabulated in Gradshteyn & Ryzhik, p. 395

$$= 2\int_0^{\infty} dt\left\{\cos at^2\cos 2bt + i\sin at^2\cos 2bt\right\}$$

$$= \sqrt{\frac{\pi}{2a}}\left\{\left[\cos\frac{b^2}{a} + \sin\frac{b^2}{a}\right] + i\left[\cos\frac{b^2}{a} - \sin\frac{b^2}{a}\right]\right\}$$

$$= \sqrt{\frac{\pi}{2a}}(1+i)e^{-i(b^2/a)}, \text{ w/ } b^2/a = \alpha^2\kappa^2, \ \underline{\underline{\alpha}} = \sqrt{\tau/4\Delta\nu}. \quad (2)$$

$\alpha$ has the dimensions of a time. Using also $(1+i)/\sqrt{2} = e^{i\frac{\pi}{4}}$, we find...

$$\boxed{\delta(\kappa) = (\alpha/\sqrt{\pi})\, e^{i\frac{\pi}{4}}\, e^{-i\alpha^2\kappa^2}}, \quad (3)$$

as the spectral fcn for the chirped signal $\exp\left\{-i\left[\nu - \frac{t}{4\alpha^2}\right]t\right\}$. Notice that the area under this curve is cnst and independent of $\alpha$, as...

$$\rightarrow \int_{-\infty}^{\infty}\delta(\kappa)d\kappa = \frac{e^{i\pi/4}}{\sqrt{\pi}}\int_{-\infty}^{\infty} e^{-ix^2}dx = 1 \quad \text{(Fresnel Integrals : G\&R, p.395)}. \quad (4)$$

Then, when $\alpha\to\infty$, $\delta(\kappa)$ of Eq. (3) goes over to a delta-fcn, since it vanishes everywhere but <u>at</u> $\kappa=0$, where it becomes large. $\alpha\to\infty$ is the <u>monochromatic</u> limit.

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(B) **2)** The transition amplitude is : $ia(\Omega) = 2\pi\int_{-\infty}^{\infty} dk\, \delta(\Omega-k)\tilde{\mathcal{E}}(k)$, where the detuning frequency $\Omega = \omega_0 - \nu$. Change variables to $\kappa = \Omega - k$, and write...

[next page]

$\phi$507 Solutions (37 cont'd)

$$\rightarrow i a(\Omega) = 2\pi \int_{-\infty}^{\infty} d\kappa\, \delta(\kappa)\, \tilde{\mathcal{E}}(\Omega-\kappa) = 2\sqrt{\pi}\,\alpha\, e^{i\frac{\pi}{4}} \int_{-\infty}^{\infty} d\kappa\, e^{-i\alpha^2\kappa^2}\, \tilde{\mathcal{E}}(\Omega-\kappa). \quad (5)$$

We've inserted $\delta(\kappa)$ of Eq. (3). If the envelope $\mathcal{E}(t)$ is Gaussian...

$$\mathcal{E}(t) = \mathcal{E}_0 e^{-(t/T)^2}$$

soll $$\tilde{\mathcal{E}}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathcal{E}(t)\, e^{i\omega t}\, dt = \frac{\mathcal{E}_0 T}{2\pi}\int_{-\infty}^{\infty} e^{-x^2+(i\omega T)x}\, dx$$ tabulated in G&R p. 307 #(3.323.2)

❘❘ $$\tilde{\mathcal{E}}(\omega) = (\mathcal{E}_0 T/2\sqrt{\pi})\, e^{-T^2\omega^2}. \quad (6)$$

Use of this envelope transform in Eq. (5) gives the transition amplitude

$$i a(\Omega) = \alpha \mathcal{E}_0 T (e^{i\pi/4}) \int_{-\infty}^{\infty} d\kappa\, e^{-i\alpha^2\kappa^2}\, e^{-T^2(\kappa-\Omega)^2}$$

tabulated in G&R

$$\rightarrow \text{or } (e^{i\pi/4})\, a(\Omega) = \alpha \mathcal{E}_0 T\, e^{-\Omega^2 T^2} \int_{-\infty}^{\infty} e^{-(T^2+i\alpha^2)\kappa^2 + (2\Omega^2 T)\kappa}\, d\kappa$$

$$= \underline{\underline{\frac{\sqrt{\pi}\,\alpha\,\mathcal{E}_0 T}{\sqrt{T^2+i\alpha^2}}\, e^{-\Omega^2 T^2 [i\alpha^2/(T^2+i\alpha^2)]}}}. \quad (7)$$

This is the transition amplitude for a coupling: $U(t,\nu) = \mathcal{E}_0 e^{-(t/T)^2} e^{-i[\nu - \frac{t}{4\alpha^2}]t}$.

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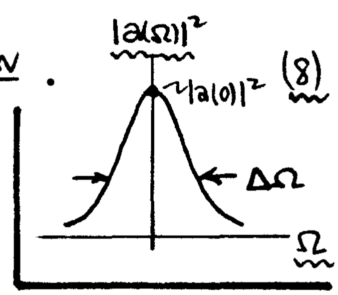
(C) **3)** The transition lineshape which follows from Eq. (7) is...

$$\rightarrow |a(\Omega)|^2 = \frac{\pi\alpha^2\mathcal{E}_0^2 T^2}{\sqrt{T^4+\alpha^4}}\, e^{-2\Omega^2T^2[\alpha^4/(T^4+\alpha^4)]}, \quad \text{w/ } \underline{\Omega = \omega_0 - \nu} \text{ detuning frequency}$$

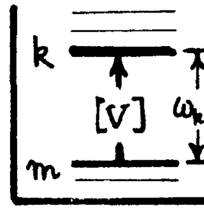
$$\text{or } \boxed{|a(\Omega)|^2 = \frac{\pi\mathcal{E}_0^2 T^2}{\sqrt{1+r^2}}\, e^{-2\Omega^2T^2/(1+r^2)}} \quad \text{w/ } \underline{\underline{r}} = \left(\frac{T}{\alpha}\right)^2 = \frac{4T^2\Delta\nu}{\tau}. \quad (8)$$




$$\Rightarrow \left.\begin{array}{l} \text{peak height}: |a(0)|^2 = \pi\mathcal{E}_0^2 T^2/\sqrt{1+r^2}, \\ \text{linewidth}: \Delta\Omega \simeq \frac{1}{T\sqrt{2}}\sqrt{1+r^2}. \end{array}\right\} (9)$$

The line is symmetric in $\Omega$; in fact it follows the symmetry of the envelope $\mathcal{E}(t)$. In the monochromatic limit, $\alpha\rightarrow\infty \Rightarrow r\rightarrow 0$, the peak $|a(0)|^2 \simeq \pi\mathcal{E}_0^2T^2$ and width $\Delta\Omega \simeq 1/T\sqrt{2}$ are determined by the envelope. For large "chirping" ($\alpha\rightarrow 0$, $r\rightarrow$ large): $|a(0)|^2 \simeq \frac{\pi}{4}(\mathcal{E}_0^2\tau/\Delta\nu)$ & $\Delta\Omega \simeq \frac{1}{\tau\sqrt{2}}(4T\Delta\nu)$, depend on the chirp parameters.

(38) [20 pts]. Analyse $\Theta(V^2)$ transitions for a pulsed harmonic perturbation.




1. For the coupling $V(x,t) = 2\hbar\Omega(x)\cos\omega t$, over $0 \leqslant t \leqslant T$, we have

(A) found the $m \to k$ transition amplitude to $\Theta(V)$ [class notes, p. tD 6]:

$$\rightarrow a_k^{(1)}(t) = \Omega_{km}\left[\frac{1-e^{i(\omega_{km}+\omega)t}}{\omega_{km}+\omega} + \frac{1-e^{i(\omega_{km}-\omega)t}}{\omega_{km}-\omega}\right], \text{ during time } V \text{ is "on".} \tag{1}$$

This amplitude corresponds to direct (single photon) transitions $m \to k$ which are resonant for absorption when $\omega \simeq \omega_{km}$.

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2. $\Theta(V^2)$ processes are governed by the amplitude $a_k^{(2)}(t)$, where [notes, p. tD13]:

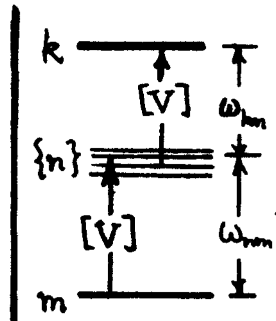
$$i\hbar a_k^{(2)}(t) = \sum_n \int_0^t d\tau\, V_{kn}(\tau)\, e^{i\omega_{kn}\tau}\left\{a_n^{(1)}(\tau)\right\} \leftarrow \text{put in } V_{kn}, \text{ and } a_n^{(1)} \text{ from Eq. (1)}$$

$$= \sum_n \int_0^t d\tau\,\left[\hbar\Omega_{kn}\left(e^{i\omega\tau}+e^{-i\omega\tau}\right)\right]e^{i\omega_{kn}\tau}\left\{\Omega_{nm}\left[\frac{1-e^{i(\omega_{nm}+\omega)\tau}}{\omega_{nm}+\omega} + \frac{1-e^{i(\omega_{nm}-\omega)\tau}}{\omega_{nm}-\omega}\right]\right\}$$

$$\xrightarrow{\text{or}} i a_k^{(2)}(t) = \sum_n \Omega_{kn}\Omega_{nm}\int_0^t d\tau\left[e^{i(\omega_{kn}+\omega)\tau} + e^{i(\omega_{kn}-\omega)\tau}\right]\cdot\left[\frac{1-e^{i(\omega_{nm}+\omega)\tau}}{(\omega_{nm}+\omega)} + \frac{1-e^{i(\omega_{nm}-\omega)\tau}}{(\omega_{nm}-\omega)}\right]. \tag{2}$$

The integrals are straightforward, but numerous. Since $\omega_{kn}+\omega_{nm} = \omega_{km}$, etc. some "cancellations" occur. Also, there are cross terms that produce oscillations $e^{i\omega_{km}\tau}$ which are non-resonant; they can be dropped immediately. After such arithmetic, we obtain...

$$a_k^{(2)}(t) \simeq \sum_n \frac{\Omega_{kn}\Omega_{nm}}{\omega_{nm}+\omega}\left[\frac{1-e^{i(\omega_{kn}+\omega)t}}{\omega_{kn}+\omega} - \frac{1-e^{i(\omega_{km}+2\omega)t}}{\omega_{km}+2\omega} + \frac{1-e^{i(\omega_{kn}-\omega)t}}{\omega_{kn}-\omega}\right] +$$
$$+ \sum_n \frac{\Omega_{kn}\Omega_{nm}}{\omega_{nm}-\omega}\left[\frac{1-e^{i(\omega_{kn}-\omega)t}}{\omega_{kn}-\omega}\,\text{①} - \frac{1-e^{i(\omega_{km}-2\omega)t}}{\omega_{km}-2\omega}\,\text{②} + \frac{1-e^{i(\omega_{kn}+\omega)t}}{\omega_{kn}+\omega}\right]. \tag{3}$$




These terms govern two-photon transitions: $m \to \{n\}$, $\{n\} \to k$, thru a set of intermediate states $\{n\}$ as depicted at right. Photons at frequencies $\omega \simeq \omega_{nm}$ and $\omega \simeq \omega_{kn}$ are required for resonance. If $\omega \simeq \frac{1}{2}\omega_{km}$, only terms ① & ② can be doubly resonant this way.

3. We retain terms ① & ② in Eq. (3) for a possible resonant absorption @ $\omega \simeq \frac{1}{2}\omega_{km}$.

(B) First, rewrite them in terms of the following notation...

$$\left.\begin{array}{l} \underline{\underline{\omega_0}} = \frac{1}{2}\omega_{km},\ ^{\text{so}}\!/\!/\ \omega_{km} - 2\omega = -2\Delta\omega,\ ^{\text{w}}\!/\!/\ \underline{\underline{\Delta\omega}} = \omega - \omega_0 \text{ (detuning)}; \\ \underline{\underline{\delta_{nm}}} = \omega - \omega_{nm},\ ^{\text{so}}\!/\!/\ \omega_{kn} - \omega = \omega_{km} - \omega - \omega_{nm} = -2\Delta\omega + \delta_{nm}; \end{array}\right\} \quad (4)$$

$$^{\text{so}}\!/\!/\ \boxed{a_k^{(2)}(t) \simeq \sum_n \frac{\Omega_{kn}\Omega_{nm}}{\delta_{nm}}\left[\frac{1-e^{-i(2\Delta\omega-\delta_{nm})t}}{2\Delta\omega-\delta_{nm}} - \frac{1-e^{-i(2\Delta\omega)t}}{2\Delta\omega}\right]}. \quad (5)$$

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(C) 4. Both $\Delta\omega$ and $\delta_{nm}$ can $\to 0$, $\Delta\omega$ by tuning, and $\delta_{nm}$ by an "accident" where some energy $\omega_{nm}$ happens to match $\omega$. Consider the following cases.

I. Both $\delta_{nm}$ and $\Delta\omega \to 0$. Use: $\frac{1}{k}(1-e^{-ikt}) \simeq it + \frac{1}{2}kt^2 + \ldots$, as $k \to 0$:

$$^{\text{so}}\!/\!/ \rightarrow a_k^{(2)}(t) \simeq -\frac{1}{2}\Big(\tilde{\sum_n}\Omega_{kn}\Omega_{nm}\Big)t^2, \quad (6)$$

$\tilde{\sum_n}$ means a sum restricted to those values of $n$ where $\delta_{nm} = \omega - \omega_{nm} \simeq 0$.

II. $\delta_{nm} \to 0$, but $\Delta\omega \neq 0$. If we define $\epsilon = \frac{\delta_{nm}}{2\Delta\omega} \ll 1$, the [ ] in Eq. (5) is

$$[\text{Eq}(5)] = \frac{1}{2\Delta\omega}\left[\frac{1}{1-\epsilon}\left\{1 - e^{-2it\Delta\omega}(e^{2i\epsilon t\Delta\omega})\right\} - \left\{1 - e^{-2it\Delta\omega}\right\}\right]$$

$$\simeq \frac{\epsilon}{2\Delta\omega}\left[1 - (1 + 2it\Delta\omega)e^{-2it\Delta\omega}\right], \text{ to 1st order in } \epsilon. \quad (7)$$

$$^{\text{so}}\!/\!/ \rightarrow a_k^{(2)}(t) \simeq +\frac{1}{4(\Delta\omega)^2}\Big(\tilde{\sum_n}\Omega_{kn}\Omega_{nm}\Big)\left[1 - (1 + 2it\Delta\omega)e^{-2it\Delta\omega}\right]. \quad (8)$$

III. $\delta_{nm} \neq 0$, but $\Delta\omega \to 0$. If $|\Delta\omega| \ll |\delta_{nm}|$, and $t\Delta\omega \ll 1$, then...

$$[\text{Eq.}(5)] \simeq \frac{1-(1-2it\Delta\omega)e^{it\delta_{nm}}}{0-\delta_{nm}} - \frac{1-(1-2it\Delta\omega)}{2\Delta\omega} \simeq -\left(\frac{1}{\delta_{nm}} + it\right) \quad (9)$$

This assumes $t\delta_{nm}$ is large enough so that $e^{it\delta_{nm}}$ averages to zero. Then

$$\rightarrow a_k^{(2)}(t) \simeq (-)\sum_n \frac{\Omega_{kn}\Omega_{nm}}{\delta_{nm}^2}(1 + it\delta_{nm}). \quad (10)$$

Comparing (6) & (10), clearly the resonant behavior depends critically on the $\delta_{nm}$.