

EE 236
11/29/04
①

Nonlinear Optics

QM: Yariv 3.17

Prop. Yariv chapter 13

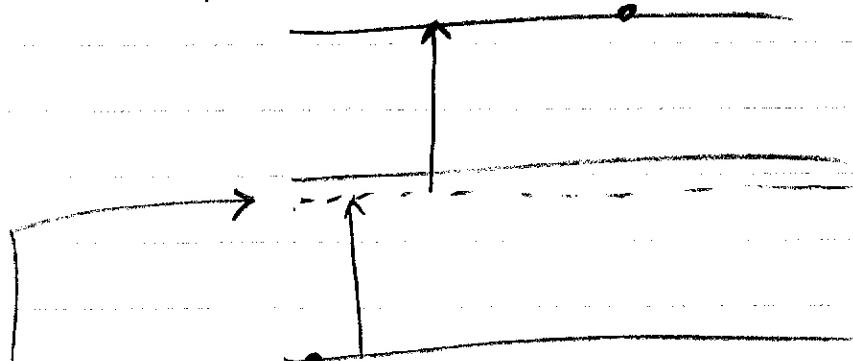
Uses: sum + difference frequency generation
& frequency doubling
pump + probe for ultrahigh speed
experiments

Parametric amplification

Raman + Brillouin scattering

(stimulated Raman amps ...)

We discussed second order processes in a handwaving way using the energy diagram:



the dotted line represents a small occupation of the nearby level with an offset oscillation in phase \rightarrow resulting in the need to coherently make another transition

(2)

This can be done analytically by introducing the time evolution operator

The time evolution operator is sort of intermediate between the Schrödinger picture and the Heisenberg picture (where the operators have the time dependence)

The def. of the time propagation operator is:

$$|\Psi(t_b)\rangle = \underbrace{\hat{U}(t_b, t_a)}_{\text{↑}} |\Psi(t_a)\rangle$$

we can then plug this into the Schrödinger eq.

$$i\hbar \frac{\partial}{\partial t_b} |\Psi(t_b)\rangle = \hat{H} |\Psi(t_b)\rangle$$

$$i\hbar \frac{\partial}{\partial t_b} \hat{U}(t_b, t_a) |\Psi(t_a)\rangle = \hat{H} \hat{U}(t_b, t_a) |\Psi(t_a)\rangle$$

Since this holds for any state $|\Psi(t_a)\rangle$ we can write the operator equation

$$i\hbar \frac{\partial}{\partial t_b} \hat{U}(t_b, t_a) = \hat{H} \hat{U}(t_b, t_a)$$

(3)

Since we can expand $\hat{\psi}(t=0)$ over the eigenstates of \hat{H}

$$\psi(t=0) = \sum_m A_m |\phi_m\rangle$$

we have

$$\psi(t) = \sum_m A_m e^{i\omega_m t} |\phi_m\rangle$$

and

$$U(t_b, t_a) = \sum_m |\phi_m\rangle e^{i\omega_m(t_b-t_a)} \langle \phi_m|$$

Now use perturbation:

$$H = H_0 + V(t)$$

Since $U(t_b, t_a)$ is a unitary operator (since $|\psi(t)\rangle$ remain normalized)

$$1 = \langle \psi(t_b) | \psi(t_b) \rangle =$$

$$\langle \hat{U}(t_b, t_a) \psi(t_a) | \hat{U}(t_b, t_a) \psi(t_a) \rangle$$

$$= \langle \psi(t_a) | \hat{U}^+(t_b) \hat{U}(t_b, t_a) | \psi(t_a) \rangle$$

$$\Rightarrow \hat{U}^+(t_b, t_a) \hat{U}(t_b, t_a) = I$$

(works for any operator which maintains normalization)

(4)

from the definition of \hat{U} :

$$\hat{U}(t_b, t_a) \hat{U}(t_a, t_c) = \hat{U}(t_b, t_c)$$

$$\text{therefore } \hat{U}(t_a, t_b) = \hat{U}^+(t_b, t_a)$$

if we now define $\hat{U}^{(0)}(t_a, t_b)$
to be the solution to

$$i\hbar \frac{\partial}{\partial t_b} \hat{U}(t_b, t_a) = \hat{H}_0 \hat{U}^{(0)}(t_b, t_a)$$

and apply schrödinger's equation to
the Hamiltonian $\hat{H} = \hat{H}_0 + \hat{V}(t)$

we have

$$i\hbar \frac{\partial}{\partial t_b} \hat{U}(t_b, t_a)$$

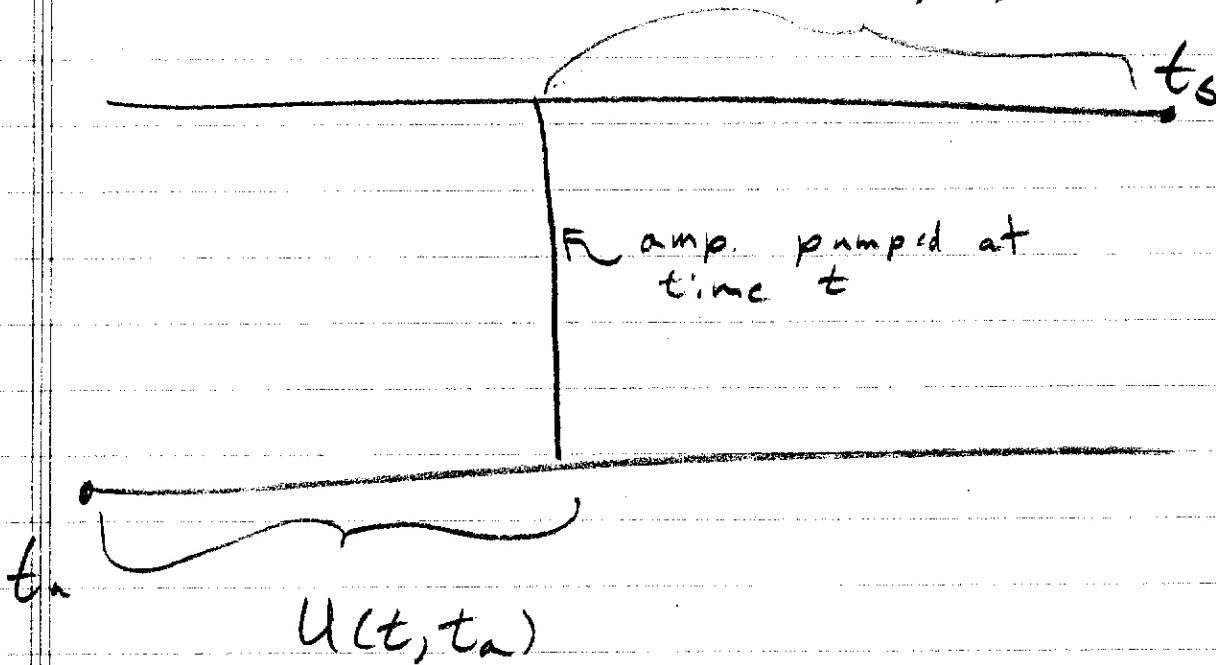
$$= \hat{H}_0 \hat{U}(t_b, t_a) + \hat{V}(t_b) \hat{U}(t_b, t_a)$$

this can be interpreted as the
time rate of change of $\hat{U}(t_b, t_a)$ is
equal to the time rate of change of
the unperturbed state plus the amplitude
pumped from the other states by $\hat{V}(t_b)$

(5)

We then want to integrate over all possible paths for amplitude to get from one state to another

$$U(t, t_0)$$



the question is, what time propagator do we use from $t_a \rightarrow t$ & $t \rightarrow t_0$?

If we use $U^{(0)}$ for both, then we only allow the perturbation to act once.

If we use U for both, we double count, \rightarrow so we use $U^{(0)}$ for one, and U for the other.

We must also account for the amp. which does not get pumped by $V(t)$

(6)

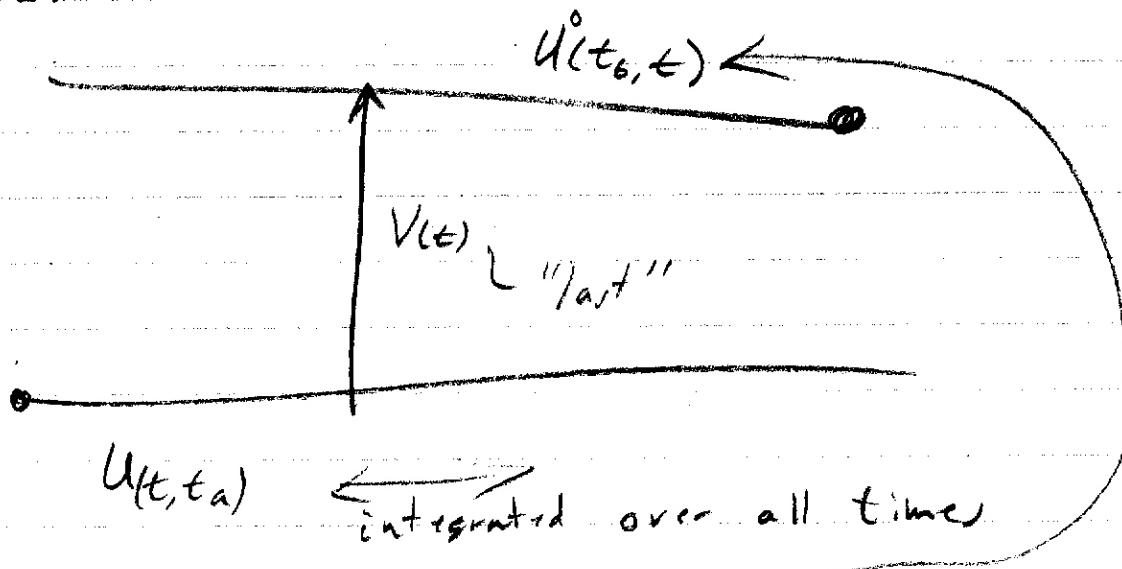
we then have:

$$\hat{U}(t_s, t_a) = U^{(0)}(t_s, t_a)$$

$$- \frac{i}{\hbar} \int_{t_a}^{t_s} U^{(0)}(t_s, t) V(t) \hat{U}(t, t_a) dt$$

notice that (this term is the perturbed time propagation operator.

This corresponds to the sum of probability amplitude along all paths



Note that if $U(t_s, t)$ appeared instead of $U^{(0)}(t_s, t)$ we would be double counting the paths

(7)

If the perturbation $V(t)$ is small, then $U^{(0)}(t, t_0)$ should be close to $U(t, t_0)$ so if we plug in the unperturbed $U^{(0)}(t, t_0)$ into the integral, we should get a better approximation to $U(t_0, t_0)$.

Each time we do that, we get a "correction" to U

$$U(t_0, t_0) = U^{(0)}(t_0, t_0) + U''(t_0, t_0) + U^{(2)}(t_0, t_0)$$

where $U^{(0)}(t_0, t_0) = \sum_m | \phi_m \rangle e^{i (U_0 t_0 - E_m)} \langle \phi_m |$

$$U''(t_0, t_0) = \left(-\frac{i}{\hbar}\right) \int_{t_0}^{t_0} U^{(0)}(t_0, t_1) V(t_1) U^{(0)}(t_1, t_0) dt_1$$

$$U^{(2)}(t_0, t_0) = \left(-\frac{i}{\hbar}\right)^2 \int_{t_0}^{t_0} \int_{t_0}^{t_0} U^{(0)}(t_0, t_1) V(t_1) U^{(0)}(t_1, t_2) V(t_2) U^{(0)}(t_2, t_0) dt_1 dt_2$$

Each successive correction takes into account a differential bit of amplitude which is jumped by $V(t_n)$, with the propagation of amplitude between those times propagating by $U^{(0)}$ from the unperturbed Hamiltonian.

(8)

If we take the perturbation to be a sum of sinusoidal perturbations:

$$V(t) = M \left(\frac{E_1}{2} e^{i\omega_1 t} + \frac{E_2}{2} e^{i\omega_2 t} + \dots \right)$$

We can then write (if start in state $|\phi_n\rangle$)

$$\psi^{(0)}(t) = \psi^{(0)}(t, 0) = e^{-i\omega_n(t-t_0)} |\phi_n\rangle$$

$$\psi'(t) = \psi''(t, 0) = -\frac{i}{\hbar} \int_{t_0}^t$$

$$\left(\sum_l |\phi_l\rangle e^{i\omega_l(t-t_0)} \langle \phi_l| \right) V(t_0)$$

$$\left(\sum_k |\phi_k\rangle e^{i\omega_k(t-t_0)} \langle \phi_k| \right) dt, |\phi_n\rangle$$

and so on.

Now, there are obviously a large number of terms, from all of the frequencies and states which could be involved.

(9)

However, each of the terms will involve integrations over sinusoidal elements which will average to a negligible amount except for those cases where the net oscillation is small,

When the integration is carried out, we get a factor in the denominator for each action of $V(t)$,

$$\Psi^{(1)}_t = \frac{1}{2\pi} \sum_m (H)_{mn} E_1^* e^{-i(\omega_m t)} | \phi_m \rangle \\ \cdot \frac{e^{i(\omega_{mn}-\omega_1)t} - e^{-i(\omega_{mn}-\omega_1)t}}{(\omega_{mn} - \omega_1)}$$

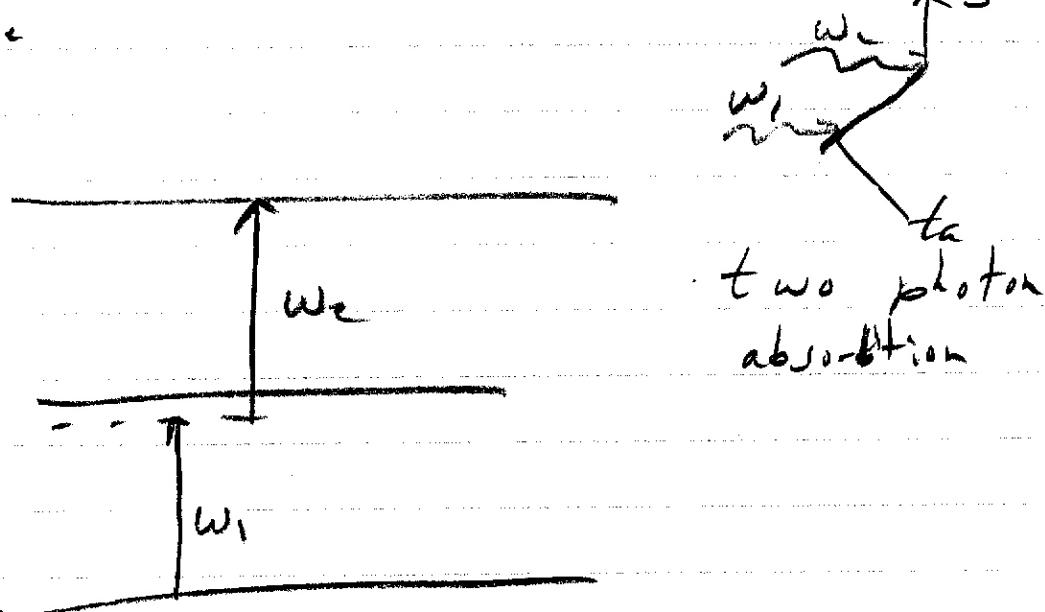
Similar to that for time dependant perturbation

$$\Psi^{(2)}_{-\omega_1 - \omega_2} = \sum_m \sum_s \left(\frac{1}{\pi} \right) \frac{E_1^* E_2^*}{q} (H_1)_{mn} (H_2)_{sm} \\ \cdot \frac{e^{i(\omega_m - \omega_1 - \omega_2)t}}{(\omega_{mn} - \omega_1)(\omega_{sm} - \omega_1 - \omega_2)}$$

Of course, the zeros in the denominators are due to our

(10)

neglect of such factors such as spontaneous emission, in general there will be a damping of the resonance corresponding to the τ_{loss} time



the terms in the denominator will be those corresponding to energy conservation at each interaction

$$\Psi_{-w_1, w_2}^{(2)} = \sum_n \sum_s \left(\frac{1}{\pi} \right)^L E_i^* E_s$$

$$? (M_1)_{nn} (M_2)_{sm} e^{i(w_n - w_1 + w_2)t}$$

each interaction $(w_{nn} - w_1 - i\delta)(w_{sm} - w_1 + w_2 - i\tau)$



(11)

looking at the interaction
of the E + M fields, for
a three photon interaction,
we get a polarization

$$P_i^{\omega_3} = 2 \text{disk} \frac{\omega_3 = \omega_1 + \omega_2}{E_i E_K} E_i^{\omega_1} E_K^{\omega_2}$$

The symmetry restrictions on
the nonlinear susceptibility are
similar to those for the Electro-
optic coefficient, I.e. the
3-photon term is zero for
centrosymmetric crystals.

Other considerations:

— Since the nonlinear term is
proportional to $E_1 \cdot E_2$, it is
necessary to have a strongly focused
beam to observe nonlinearities.

If a polarization field $\vec{P}(r, t)$ is
produced, it will not couple
energy out unless it is matched
to a propagating field