– RAMAN MECHANISMS

2.1 — PHENOMENOLOGICAL THEORY OF GENERATION

• TRANSPARENT MEDIA

$$U = \frac{1}{2}\chi E_0^2(\vec{r},t) + \frac{1}{8\pi}\underbrace{E_0^2(\vec{r},t)}_{\text{CLASSICAL LASER FIELD}}$$

$$\left[U = \frac{\epsilon E^2}{8\pi}\right]$$

energy density

ASSUME THAT $\chi = \chi^{(0)} + \frac{\partial\chi}{\partial Q} Q$ → PHONON FIELD

$$\boxed{\delta n = \frac{2\pi\,\delta\chi}{n}}$$

REFRACTIVE INDEX

$$\delta U = \frac{1}{2}\delta\chi E_0^2(\vec{r},t) = \frac{1}{2}\underbrace{\left(\frac{\partial\chi}{\partial Q}\right)}_{\text{CONSTANT}} Q(\vec{r},t)\, E_0^2(\vec{r},t)$$

LOCAL INTERACTION

SINCE $\delta U \propto Q \Rightarrow \mathcal{F} \propto E_0^2(\vec{r},t)$

FORCE DENSITY

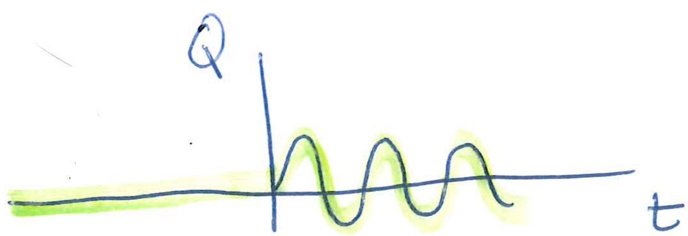
$$\Rightarrow \frac{d^2Q}{dt^2} + \Omega^2 Q = \frac{1}{2}\left(\frac{\partial\chi}{\partial Q}\right)|E_0^2(\vec{r},t)| \equiv \vec{F}(\vec{r},t)$$

$\cancel{\frac{1}{2}\frac{\partial\chi}{\partial Q} E_\alpha E_\beta}$

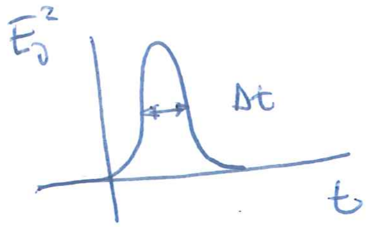
SOLUTION

$$Q(\vec{r},t) = \int_{-\infty}^{t} \frac{\sin\Omega(t-\tau)}{\Omega} F(\vec{r},t)\,d\tau$$

IF $F(\vec{r},t) \sim \delta(t)$



IMPULSIVE LIMIT



$$\Rightarrow \quad \Delta t \ll \frac{2\pi}{\Omega}$$

LATER, WE'LL DISCUSS SELECTION RULES

$$F = \sum_{\substack{\mu\nu \\ \alpha}} \underbrace{\frac{\partial \chi_{\mu\nu}}{\partial Q_\alpha}}_{\text{RAMAN TENSOR}} \mathcal{E}_\mu \mathcal{E}_\nu / 2$$

- IGNORES DEPLETION OF PUMP FIELD
- APPLIES TO TRANSPARENT MEDIA
- WHAT IS A PHONON FIELD?

- PHONON FIELDS

$$\hat{H} = \frac{1}{2} \sum_{s,\underline{l}} M_s |\dot{\vec{u}}_{s,\underline{l}}|^2 + \frac{1}{2} \sum \overleftrightarrow{G}_{s\underline{l}}^{s'\underline{l}'} \vec{u}_{s\underline{l}} \vec{u}_{s'\underline{l}'}$$

$$+ \frac{1}{2} \sum_{ij} \int \delta\chi_{ij} \mathcal{E}_i^{(0)} \mathcal{E}_j^{(0)} \, dV$$

- TRANSFORM INTO NORMAL MODES:

$$\hat{H} = \frac{1}{2} \left\{ \sum_{\alpha\vec{q}} |\dot{Q}_{\alpha,\vec{q}}|^2 + \Omega^2_{\alpha\vec{q}} Q^2_{\alpha\vec{q}} \right\} + \text{INTERACTION WITH FIELD}$$

$$Q_{\vec{q}} = \frac{1}{\sqrt{V}} \int \underbrace{Q(\vec{r})} e^{i\vec{q}\cdot\vec{r}} \, d^3r$$

FIELD
NOT A
NORMAL COORDINATE

$\Big\}$ ONE BRANCH

$$Q(\vec{r}) = \frac{1}{\sqrt{V}} \sum_{\vec{q}'} Q_{\vec{q}'} \, e^{i\vec{q}'\cdot\vec{r}}$$

$$\hat{H} = \frac{1}{2} \int |\dot{Q}(\vec{r})|^2 \, dV + \frac{1}{2V} \sum_{\vec{q}} \Omega_q^2 \int e^{i\vec{q}\cdot(\vec{r}-\vec{r}')} Q(\vec{r}) Q^{+}(\vec{r}') \, d^3r \, d^3r'$$

+ INTERACTION
W/ EM FIELD

- COMPARE WITH

$$H = \frac{1}{2}\dot{Q}^2 + \frac{1}{2}\Omega^2 Q^2$$

SAME IF $\Omega_q^2 \approx \Omega_{q=0}^2 + \cancel{\cdots}$

- OK FOR OPTICAL PHONONS BECAUSE $\lambda_{LIGHT} \gg a_{CELL}$
- NOT GOOD FOR ACOUSTIC MODES, POLARITONS, ...

UNITS

$$u_{s,l} = \sqrt{\frac{V}{\underbrace{N M_s}_{\text{ION MASS}}}}\;\frac{1}{8\pi^3}\int Q_q(\vec{r})\,\underbrace{C_q^{s}}_{\text{COEFFICIENTS}}\,e^{\vec{q}\cdot(\vec{R}_l-\vec{r})}\,d^3q\,d^3r$$

Sth ION IN l CELL

IF $Q_q(\vec{r}) \equiv$ CONSTANT

$$\Longrightarrow\quad u_{s,\vec{l}} = \sqrt{\frac{V_{CELL}}{M_s}}\,C_0^{(s)}\,Q_0$$

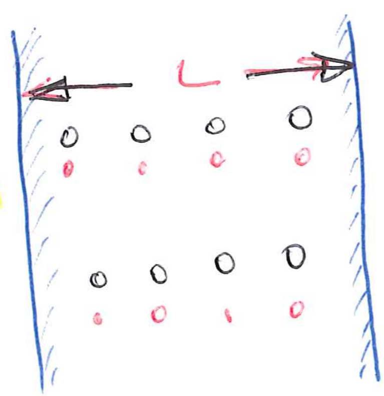
$$\frac{\partial \chi}{\partial Q} = \sum_{\substack{\text{CELL}\\(s)}} \frac{\partial \chi}{\partial u_s}\sqrt{\frac{V_{CELL}}{M_s}}\,C_0^{(s)}$$

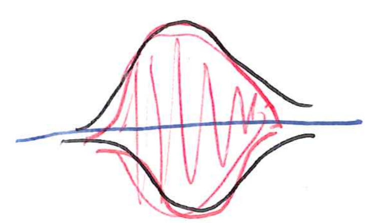
LET'S MOVE ON TO THE PROBE (DETECTION)

1. ASSUME THAT THE TIME DEPENDENCE OF $Q(\vec{r},t)$ CAN BE IGNORED –

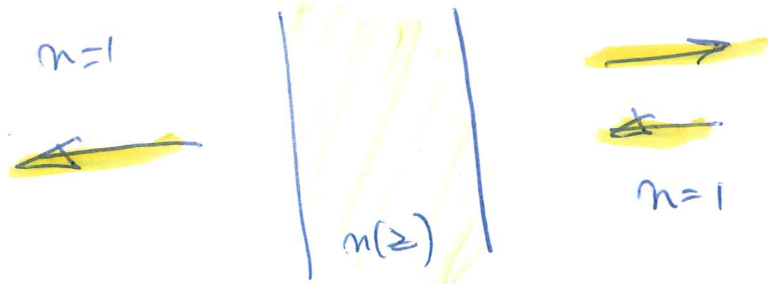
$$\frac{Ln}{c} \ll \tau_{PULSE}$$

(MORE LATER)





PROBE SEES STATIC $n(\vec{r})$

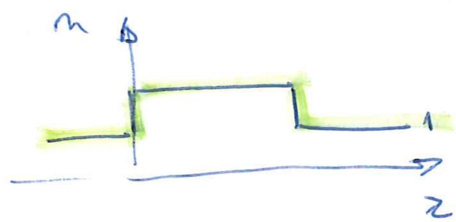


THE EQUATION TO SOLVE IS

electric field of probe

$$\left[\nabla^2 - \left\{n^2(z) + 4\pi \frac{\partial \chi}{\partial Q} Q(z)\right\} \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right] e = 0$$

$$\delta n = \frac{2\pi}{n}\delta\chi$$



TREAT THE TERM $\propto Q$ AS A PERTURBATION

LET $e_\omega(z)$ BE THE SOLUTION OF

$$\left[\nabla^2 + \frac{n^2(z)\,\omega^2}{c^2}\right] e_\omega = 0$$

INCLUDES
- INCIDENT
- REFLECTED
- TRANSMITTED

$$\int_{-\infty}^{\infty} n^2(z)\, e_\omega e_{\omega'}\, dz = \delta(\omega-\omega')$$

SEARCH FOR

$$\Psi = \sum_\omega C_\omega \, e_\omega(z) \, e^{-i\omega_0 t}$$

WHERE $C_\omega = \delta_{\omega\omega_0} + \underbrace{F(\omega)}$

PERTURBATION $\propto \dfrac{\partial \chi}{\partial Q}$

$$\Rightarrow \sum_\omega C_\omega \frac{n^2(z)}{c^2} \left(\omega_0^2 - \omega^2\right) e_\omega(z)$$

$$= -\frac{4\pi}{c^2} \omega_0^2 \frac{\partial \chi}{\partial Q} \sum_\omega Q(z) \, C_\omega \, e_\omega(z)$$

REPLACING,
MULTIPLYING * $e_{\omega'}(z)$
& INTEGRATING OVER Z

$$\frac{4\pi}{c^2} \frac{\partial \chi}{\partial Q} \omega_0^2 \int e^*_{\omega'}(z) \, Q(z) \, e_{\omega_0}(z) \, dz$$

$$= F(\omega') \frac{\left(\omega_0^2 - \omega'^2\right)}{c^2} + \frac{4\pi \omega_0^2}{c^2} \frac{\partial \chi}{\partial Q} \sum_\omega F(\omega) \int e^*_\omega Q(z) e^*_{\omega'} \, dz$$

TO LOWEST ORDER IN $\partial \chi / \partial Q$

$$\boxed{F(\omega') \simeq 4\pi \frac{\partial \chi}{\partial Q} \frac{\omega_0^2}{\omega_0^2 - \omega'^2} \int e^*_{\omega'} \, Q(z) \, e_{\omega_0} \, dz}$$

THEREFORE

$$\Psi_{\omega_0} \approx e_{\omega_0}(z) + 4\pi \frac{\partial \chi}{\partial Q}\,\omega_0^2 \sum_{\omega'} \frac{\left[\int_{\text{SLAB}} e^*_{\omega'}\, Q(z)\, e_{\omega_0}\, dz\right]}{(\omega_0^2 - \omega'^2)}\, e_{\omega'}(z) \qquad (*)$$

$$\times\, e^{-i\omega_0 t}$$

VERY USEFUL FORMULA FOR ACOUSTIC PHONONS, ACOUSTIC MAGNONS, ...

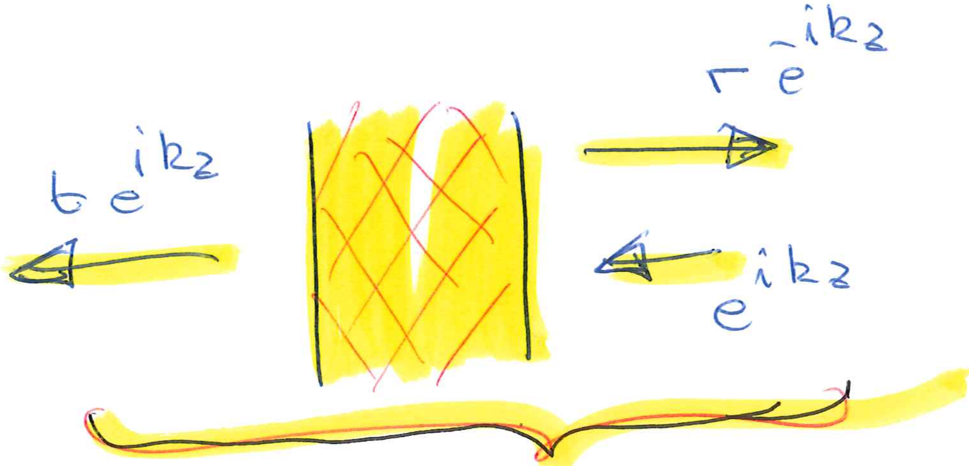
NOTE:

IF $\frac{\partial Q}{\partial z} = 0 \qquad \int_{\text{SLAB}} e^*_{\omega'} e_{\omega_0}\, dz \neq \delta_{\omega'\omega_0}$

THIS EXPRESSION IS VALID IF THE LIGHT TRANSIT TIME $\frac{Ln}{c}$ IS $\ll \tau_{PULSE}$ WHICH, IN ITSELF IS $\ll \frac{2\pi}{\Omega}$

FOR $\tau \sim 10^2$ fs & $n = 3$ $\quad L \ll 10^5$ Å

THEN, THE TIME TO REFLECT IS SIMPLY τ_{PULSE}



WHEN WE INTEGRATE $(*)$, WE NEED TO WORRY ABOUT THE POLE @ ω_0

$e_\omega(z)$

gives zero

$$\sim \sum_{\omega'} \frac{e_{\omega'}(z)}{\omega_0^2 - \omega'^2} = \frac{1}{2\omega_0} \sum_k \frac{e^{ikz} + r e^{-ikz}}{k - k_0} = \frac{1}{2\omega_0} \sum_k \frac{r e^{-ikz}}{k - k_0}$$

IF L IS LARGE, WE NEED A DIFFERENT APPROACH.

MAXWELL'S EQUATION IS

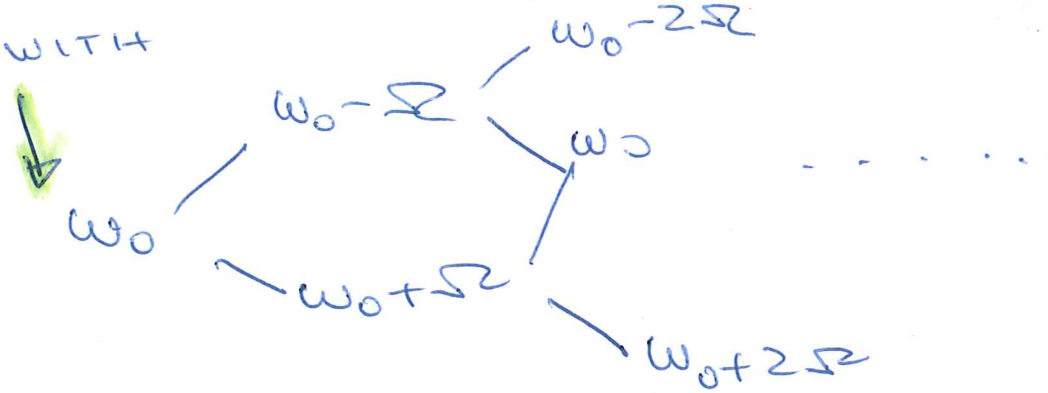
$$\vec{\nabla}\times\left(\vec{\nabla}\times\vec{E}\right) - \frac{n^2}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\vec{P}_{NL}$$

$$\vec{P} = \delta\chi\,\vec{E}$$

IF $\delta\chi$ DEPENDS ON TIME $\Rightarrow$ POLARIZATION IS NONLINEAR

CARS

START WITH



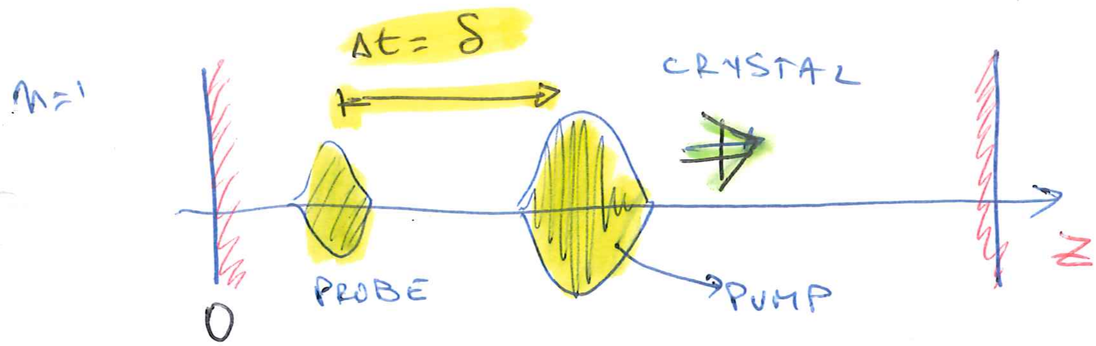
IF THE PROBE DEPLETION CAN BE IGNORED

NOTE THAT $Q \propto \frac{\partial \chi}{\partial Q}$

$$\Rightarrow \vec{P} \propto \frac{\partial \chi}{\partial Q}\, Q(\vec{r},t)\, \underbrace{e^{(0)}_{PROBE}}_{\text{UNPERTURBED}}$$

$\Rightarrow$ WE NEED TO SOLVE

$$\frac{\partial^2 e}{\partial z^2} = \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\left(n^2 e + 4\pi \frac{\partial \chi}{\partial Q}\, Q\, e^{(0)}\right)$$



$$u = t - n\frac{z}{c}$$

PUMP: $\mathcal{E}_0 \, e^{-t^2/2\tau_0^2} \cos(\omega_0 t)$

$$\Rightarrow \quad Q(u) \simeq \frac{\pi^{1/2}\tau_0}{4\Omega} |\mathcal{E}_0|^2 e^{-\Omega^2\tau_0^2/4} \frac{\partial\chi}{\partial Q} \sin \Omega u$$

(IGNORING DECAY) $\boxed{u>0}$

$$\Rightarrow \quad \frac{\partial^2 e}{\partial z^2} = \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\left(n^2 e + 4\pi \frac{\partial\chi}{\partial Q} Q(u)\, e^{(0)}(u-\delta) \right)$$

CHANGE VARIABLES
$(t,z) \to (u,z)$

$$\begin{cases} \dfrac{\partial^2}{\partial z^2} = \left(-\dfrac{n}{c}\dfrac{\partial}{\partial u} + \dfrac{\partial}{\partial z} \right)^2 \\[2ex] \dfrac{\partial^2}{\partial t^2} = \dfrac{\partial^2}{\partial u^2} \end{cases}$$

$$\Rightarrow \left[\frac{\partial^2}{\partial z^2} - \frac{2n}{c}\frac{\partial^2}{\partial z \partial u} \right] e = \frac{4\pi}{c^2}\frac{\partial\chi}{\partial Q}\frac{\partial^2}{\partial u^2}\left[Q(u)\, \bar{e}^{(0)}(u-\delta) \right]$$

EXACT SOLUTION IS

$$e = e^{(0)}(u-\delta) - z\,\frac{2\pi}{cn}\,\frac{\partial X}{\partial Q}\,\frac{\partial}{\partial u}\left[Q(u)\,e^{(0)}(u-\delta)\right]$$

(*)

GROWTHS WITH z

PUT A DETECTOR @ $z=L$ & MEASURE $|e(z=L)|^2$

$$|e(z=L)|^2 \approx |e^{(0)}|^2 - \frac{4\pi L}{cn}\,\frac{\partial X}{\partial Q}\,\times$$

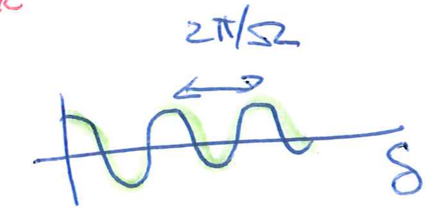
$$\times\left\{ e^{(0)}\left(t-\frac{nL}{c}-\delta\right)\left[\frac{\partial}{\partial u}\,Q(u)\,e^{(0)}(u-\delta)\right]\right\}_{z=L}$$

IF WE INTEGRATE OVER u, WE GET

$$\frac{\int |e|^2\,du}{\int |e^{(0)}|^2\,du} \approx 1 - \frac{L\sigma}{c}\cos(\Omega\delta) \quad !$$

NOT TRUE FOR PHOTON COUNTER

$$\sigma = \frac{\pi^{3/2}\tau_0}{m}\,|E_0|^2\left|\frac{\partial X}{\partial Q}\right|^2$$

1. 

2. $\propto L$
PHASE MATCHING OR WAVEVECTOR CONSERVATION

3. $\cos\Omega\delta$
ENERGY TRANSFER BETWEEN LIGHT & VIBRATION LARGEST @ $Q=0$

NEXT, GET FOURIER TRANSFORM OF (*)

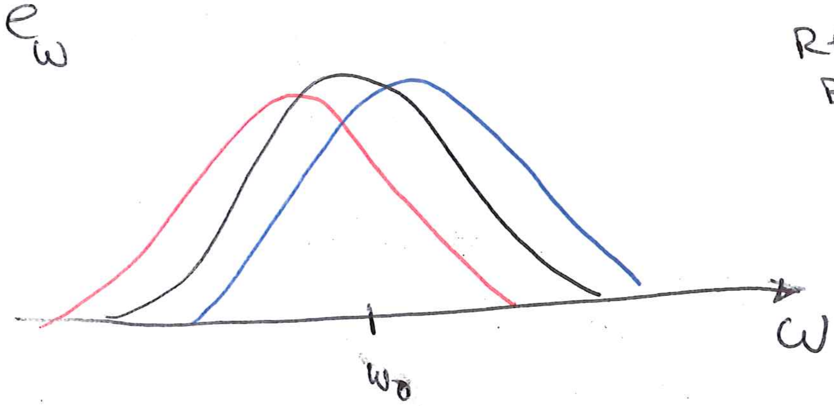
FOR $Q(u) = A \sin(\Omega u)$

& $e^{(0)}(u) = \int e^{(0)}_{\omega} \exp(i u \omega)\, d\omega$

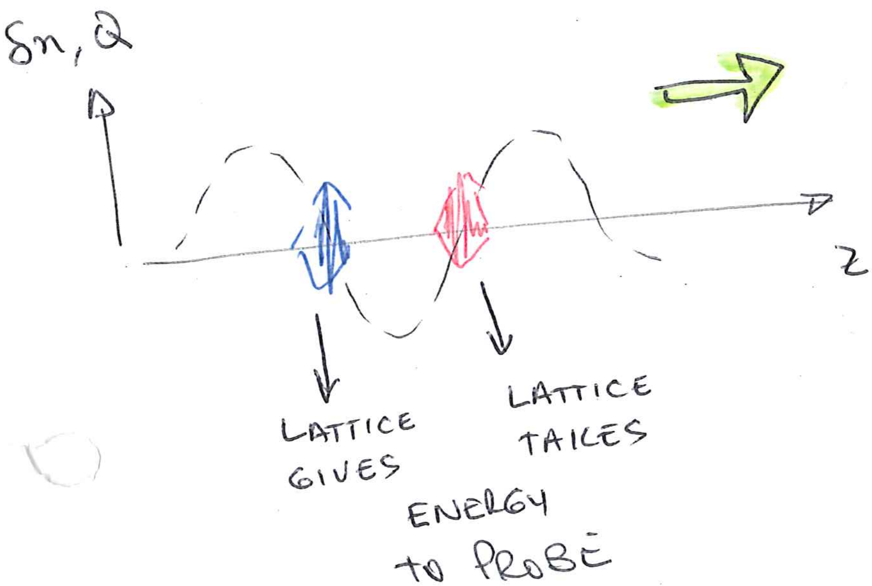
$$\Rightarrow e_{\omega} = e^{(0)}_{\omega} + \frac{\pi L A_0}{mc} \frac{\partial \chi}{\partial Q} \times$$

$$\times \left[e^{i\Omega\delta} e^{(0)}(\omega+\Omega) - e^{-i\Omega\delta} e^{(0)}(\omega-\Omega) \right]$$

Y.-X. YAN, E. B. GAMBLE JR. & KEITH A. NELSON
J. CHEM. PHYS 83, 5391 (1986) $\Leftarrow$



RED OR BLUE SHIFTED DEPENDING ON δ



FOR A SYMMETRIC PULSE

$$\frac{dI_\omega}{dQ} \approx \cos(\Omega\delta) \frac{d|e^{(0)}|^2}{d\omega}$$

$\times 2\Omega\omega$