Lecture 13. Stimulated Raman Scattering

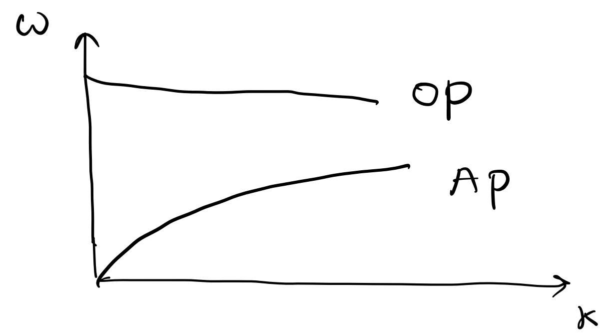
1. Stimulated Raman scattering
2. Raman gain and amplification

Recap:

Last time: Raman & Brillouin Scatterings

Raman ↙ Molecular vibration (optical phonon OP)

Brillouin ↘ Acoustic phonons (AP)

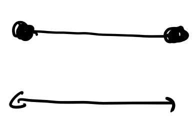


Common idea between Raman & Brillouin:

Electric field modifies quantity "q"

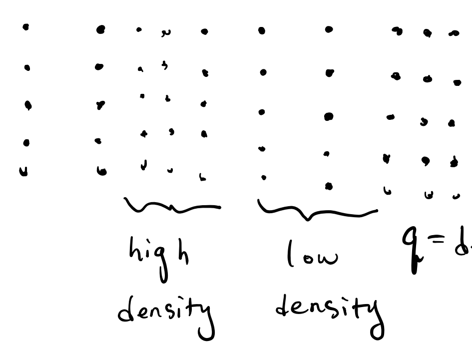
Polarization is modified by the change in q.

Raman: diatomic molecule



q = intermolecular seperation

Brillouin: acoustic wave



q = density ρ

In general, write

$$P = N \epsilon_0 \alpha E_i$$

N = number density

α = polarizabity (per atom/molecule)

The polarizability α depends on q:

$$\alpha(q) \simeq \alpha_0 + \left(\frac{\partial \alpha}{\partial q}\right)_{q_0} \tilde{q}(t) \rightarrow q_0 \cos(\Omega t)$$

$$P = \underbrace{\epsilon_0 N_0 \alpha_0 E_i}_{P^{(1)}} + \underbrace{\epsilon_0 N \left(\frac{\partial \alpha}{\partial q}\right)_{q_0} \tilde{q}(t) E_i}_{P^{(NL)}}$$

$q - q_0$ depends on E

Assume $E_i = E_0 \cos(\omega_p t)$

$$P^{(NL)} = \frac{\epsilon_0 N \cdot q_0}{2} \left(\frac{d\alpha}{dq}\right)_0 \left[\cos[(\omega_p + \Omega) t] + \cos[(\omega_p - \Omega) t]\right]$$

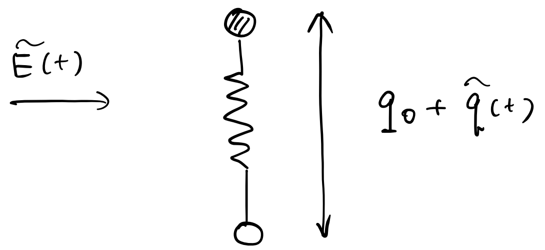
$\cos[(\omega_p + \Omega) t] \rightarrow$ Anti-Stokes wave

$\cos[(\omega_p - \Omega) t] \rightarrow$ Stokes wave

Raman: $\frac{\Omega}{2\pi} \simeq 10\,\mathrm{THz}$. lifetime of OP: 10~100 fs

Brillouin: $\frac{\Omega}{2\pi} \simeq 10\,\mathrm{GHz}$. lifetime of AP: ~10 ns

1. Stimulated Raman Scattering (SRS)



Classical harmonic oscillator model.

$$\frac{d^2\tilde{q}}{dt^2} + 2\gamma\frac{d\tilde{q}}{dt} + \Omega^2\tilde{q} = \frac{\tilde{F}(t)}{m} \qquad (1)$$

how to calculate this?

Key assumption: $\tilde{\alpha}(t) = \alpha_0 + \left(\frac{\partial\alpha}{\partial q}\right)_0 \tilde{q}(t)$

$\Rightarrow \tilde{n}(t) = \sqrt{\tilde{\epsilon}(t)} = (1 + N\tilde{\alpha}(t))^{1/2}$ (index modulation)

With incident field $\tilde{E}(z,t)$, induced dipole moment of a molecule located at z:

$$\tilde{p}(z,t) = \epsilon_0 \alpha \tilde{E}(z,t)$$

Energy of oscillating dipole:

$$W = \underbrace{\frac{1}{2}\langle \tilde{p}(z,t)\cdot\tilde{E}(z,t)\rangle}_{\text{time average over an optical period.}} = \frac{1}{2}\epsilon_0\alpha\langle\tilde{E}^2(z,t)\rangle$$

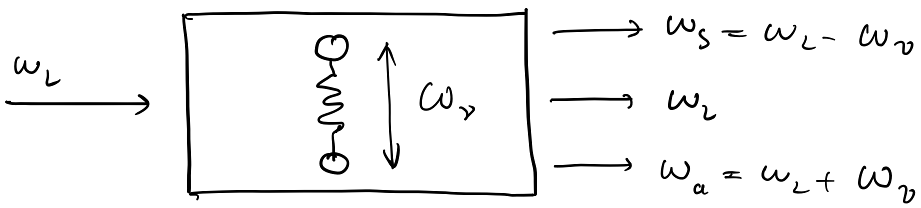
Force exerted by optical field:

$$\tilde{F} = \frac{dW}{dq} = \frac{\epsilon_0}{2}\left(\frac{d\alpha}{dq}\right)_0 \langle \tilde{E}^2(z,t) \rangle$$

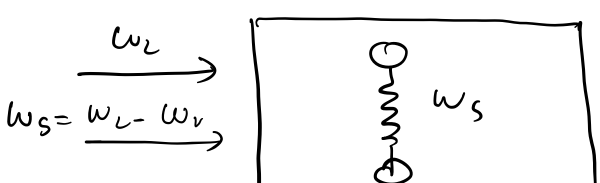
If $E = A_1 e^{i\omega_1 t} + A_2 e^{i\omega_2 t}$, $\tilde{F}$ has $(\omega_1 - \omega_2)$ term, which is the beat note!

Physical picture of SRS

① Spontaneous RS:



② Stimulated RS



1. Stokes field at $\omega_S = \omega_L - \Omega$ can beat with ω_L to produce a modulation force $\tilde{F}(t) = F_0 \cos(\omega_L - \omega_S)t = F_0 \cos(\omega_\nu t)$

2. Such a modulation coherently excites the molecular oscillation at $\omega_v = \omega_L - \omega_S$.

3. Process ① and ② reinforce each other.

Specifically, ② leads to more molecular vibration, and because of ①, stronger Stokes field, which in turn leads to stronger molecular vibration.

Input optical field:

$$\tilde{E}(z,t) = A_L e^{i(k_L z - \omega_L t)} + A_S e^{i(k_S z - \omega_S t)} + c.c.$$

$$\Rightarrow \tilde{F}(z,t) = \epsilon_0 \left(\frac{\partial \alpha}{\partial q}\right)_0 \left[A_L A_S^* e^{i(kz - \Omega t)} + c.c.\right]$$

where $k = k_L - k_S$, $\Omega = \omega_L - \omega_S$ — (generic case)

With this $\tilde{F}(z,t)$, ① has a solution in the form:

$$\tilde{q} = q(\Omega) e^{i(kz - \Omega t)} + c.c.$$

insert $\tilde{q}$ and $\tilde{F}$ into ①, we get:

$$-\omega^2 q(\Omega) - 2i\omega\gamma q(\Omega) + \omega_v^2 q(\Omega) = \frac{\epsilon_0}{m}\left(\frac{\partial \alpha}{\partial q}\right)_0 A_L A_S^*$$

Amplitude of vibration:

$$q(\Omega) = \frac{(\epsilon_0/m)(\partial\alpha/\partial q)_0 A_L \cdot A_S^*}{\omega_v^2 - \Omega^2 - 2i\Omega\gamma} \qquad (2)$$

Polarization of medium:

$$\tilde{P}(z,t) = N.\tilde{p}(z,t) = \epsilon_0 N.\tilde{\alpha}(z,t)\,\tilde{E}(z,t)$$

$$= \epsilon_0 N\left[\alpha_0 + \underbrace{\left(\frac{\partial\alpha}{\partial q}\right)_0 \tilde{q}(z,t)\Big]\tilde{E}(z,t)}_{\text{NL part}},$$

$$\tilde{P}^{NL}(z,t) = \epsilon_0 N\left(\frac{\partial\alpha}{\partial q}\right)_0 \left[q(\Omega)e^{i(Kz-\Omega t)} + c.c.\right]\left[A_L e^{i(k_L z - \omega_L t)} + A_S e^{i(k_S z - \omega_S t)} + c.c.\right]$$

Stokes polarization:

$$P_S^{NL}(z,t) = P(\omega_S)e^{-i\omega_S t} + c.c.$$

$$P(\omega_S) = N\epsilon_0\left(\frac{\partial\alpha}{\partial q}\right)_0 q^*(\Omega)\,A_L e^{ik_S z}$$

By introducing (2).

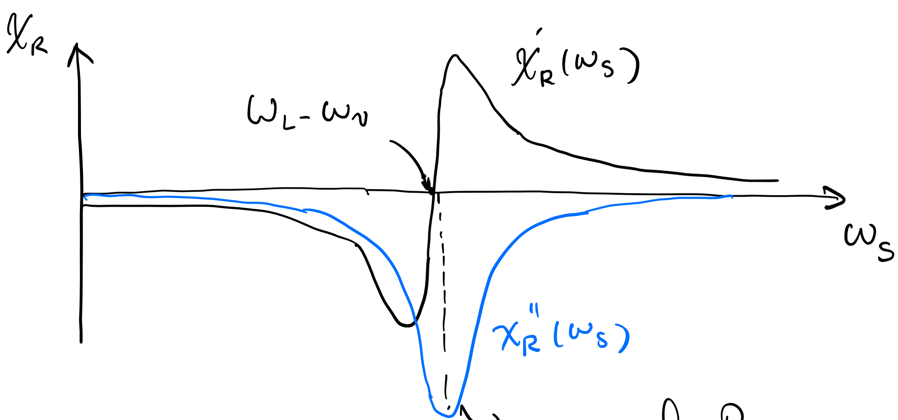
$$P(\omega_S) = \frac{(\epsilon_0^2 N/m)(\partial\alpha/\partial q)_0^2\,|A_L|^2 A_S}{\omega_v^2 - \Omega^2 + 2i\Omega\gamma}\,e^{ik_S z}$$

Define Raman susceptibility:

$$P(\omega_s) = 6\epsilon_0 \chi_R(\omega_s)\, |A_L|^2 \cdot A_s e^{ik_s z}.$$

$\chi^{(3)}(\omega_s; \omega_s, \omega_L, -\omega_L)$

$$\chi_R(\omega_s) = \frac{\epsilon_0 (N/6m)(\partial\alpha/\partial q)_0^2}{\omega_v^2 - (\omega_L-\omega_s)^2 + 2i(\omega_L-\omega_s)\gamma} = \chi_R'(\omega_s) + i\chi_R''(\omega_s)$$



around Raman resonance

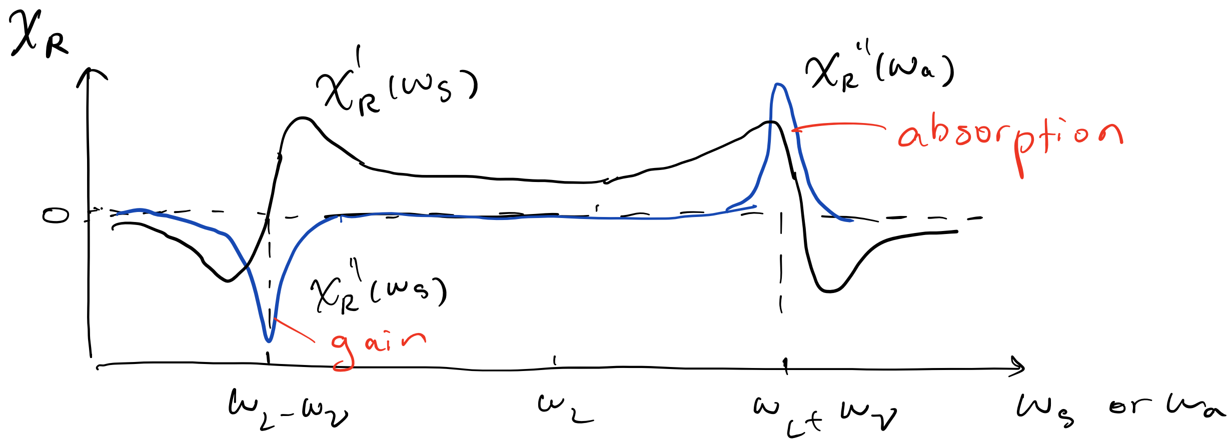
$\chi_R''(\omega_s) < 0$, $\rightarrow$ gain of Stokes wave

For anti-Stokes, just replace ω_s by ω_a

$$\chi_R(\omega_a) = \frac{\epsilon_0 (N/6m)(\partial\alpha/\partial q)_0^2}{\omega_v^2 - (\omega_L-\omega_a)^2 + 2i(\omega_L-\omega_a)\gamma}$$

Since $\omega_L - \omega_s = -(\omega_L - \omega_a)$

$$\Rightarrow \chi_R(\omega_a) = \chi_R(\omega_s)^*$$



2. Raman gain and amplifier

Input: $E = A_s e^{i(k_s z - \omega_s t)} + c.c.$

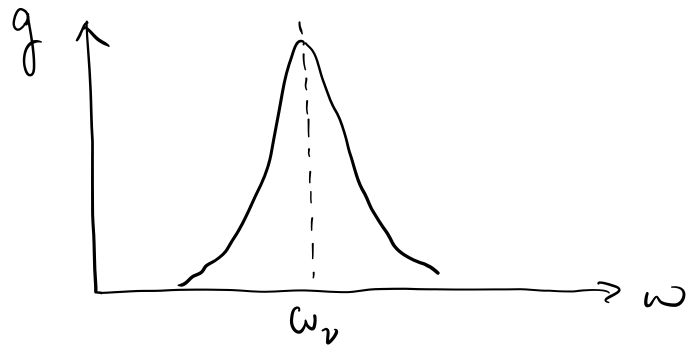
CAE: $$\frac{\partial A_s}{\partial z} = \frac{i\,\omega_s^2 \chi_R |A_L|^2}{2 k_s c^2} \cdot A_s$$

$$\Rightarrow A_s(z) = A_s(0)\, e^{\frac{i\omega_s^2 \chi_R |A_L|^2}{2k_s c^2} z}$$

χ_R has a negative imag. part $\Rightarrow$ exp. growth in A_s

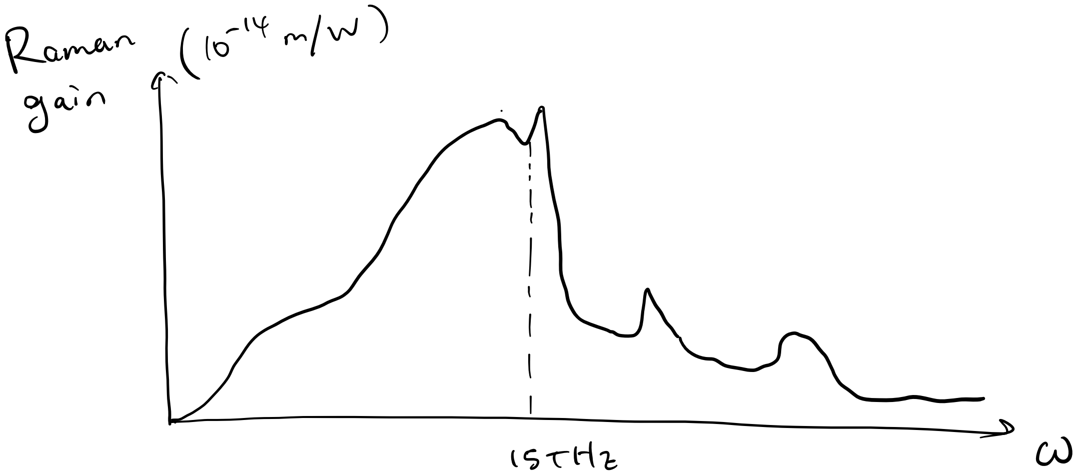
Gain coefficient: $$g = -\frac{\omega_s^2}{2k_s c^2}\,\mathrm{Im}(\chi_R)$$

(Raman-Stokes amplification)



In real materials, multiple op modes.

Eg. Silica fiber



Comments:

· phase matching is atomatically satisfied

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