Lecture 12. Raman and Brillouin Scattering

① Lattice vibration

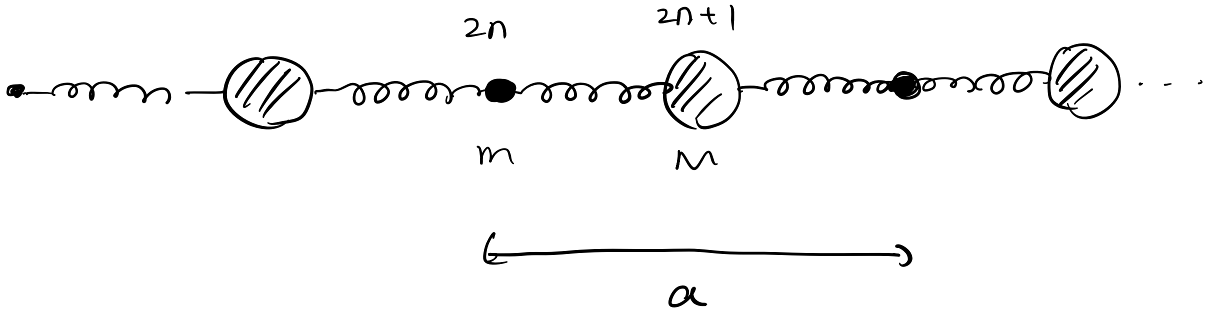
② Spontaneous light scattering

③ Spontaneous / Stimulated Raman scattering

④ Spontaneous / Stimulated Brillouin scattering

1. Lattice vibration

Model: 1-D Atomic chain. (Kittel. Solid-state physics)



Newton's second law:

spring const.

$$m \frac{d^2 q_{2n}}{dt^2} = \beta(q_{2n+1} - q_{2n}) - \beta(q_{2n-1} - q_{2n}) \quad ①$$

$$= \beta(q_{2n+1} + q_{2n-1} - 2q_{2n})$$

$$M \cdot \frac{d^2 q_{2n+1}}{dt^2} = \beta(q_{2n+2} - q_{2n+1}) + \beta(q_{2n} - q_{2n+1}) \quad ②$$

$$= \beta(q_{2n+2} + q_{2n} - 2q_{2n+1})$$

Solution of ①. ② are in the form:

$$q_{2n,k} = \xi_k e^{i(\omega t + 2nka)}$$

$$q_{2n+1,k} = \eta_k e^{i(\omega t + (2n+1)ka)}$$

plug in ①. ②, we get

$$\begin{cases} -\omega^2 m \xi_k = \beta \eta_k (e^{ika} + e^{-ika}) - 2\beta \xi_k \\ -\omega^2 M \eta_k = \beta \xi_k (e^{ika} + e^{-ika}) - 2\beta \eta_k \end{cases}$$

$$\Rightarrow (2\beta - m\omega^2)(2\beta - M\omega^2) - 4\beta^2 \cos^2(ka) = 0$$

$$\Rightarrow \omega^2 = \beta\left(\frac{1}{m} + \frac{1}{M}\right) \pm \beta \cdot \sqrt{\left(\frac{1}{m} + \frac{1}{M}\right)^2 - \frac{4\sin^2(ka)}{Mm}}$$

when $ka \ll 1$,

$$\omega^2 = \beta\left(\frac{1}{m} + \frac{1}{M}\right) \pm \beta \cdot \left(\frac{1}{m} + \frac{1}{M}\right) \cdot \left[1 - \frac{2(ka)^2}{Mm\left(\frac{1}{m} + \frac{1}{M}\right)^2}\right]$$

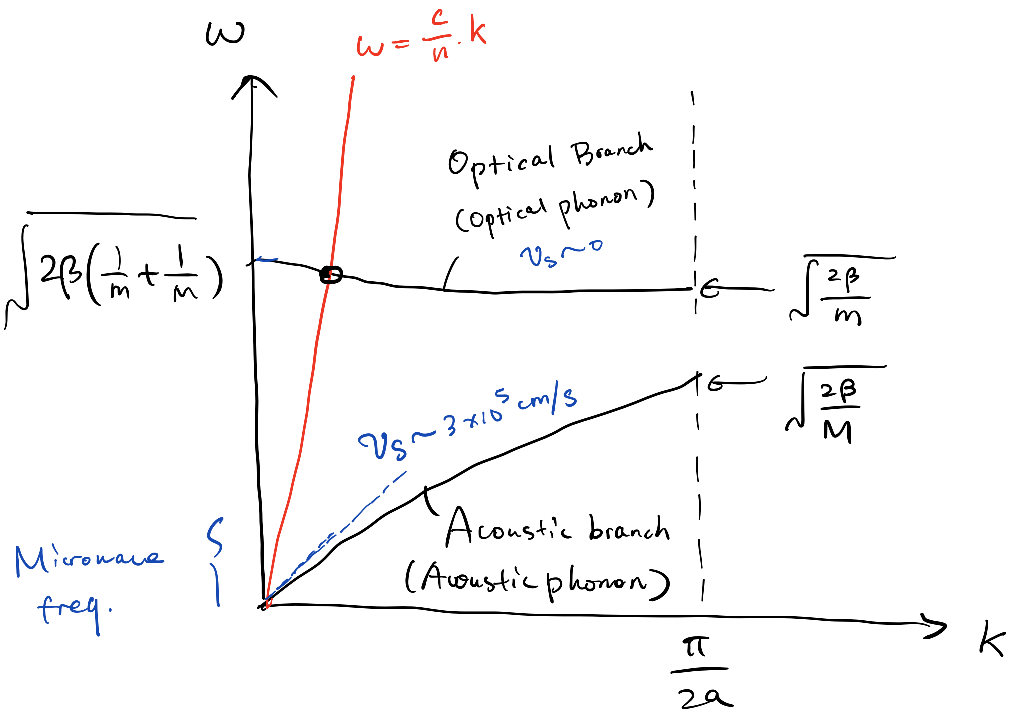
$$\Rightarrow \omega_1 = \sqrt{2\beta\left(\frac{1}{m} + \frac{1}{M}\right)}$$

$$\omega_2 = \sqrt{\frac{2\beta}{M+m}} \cdot ka$$

When $ka = \frac{\pi}{2}$,

$$\omega_1 = \sqrt{\frac{2\beta}{m}}, \quad \omega_2 = \sqrt{\frac{2\beta}{M}}$$

Dispersion diagram:



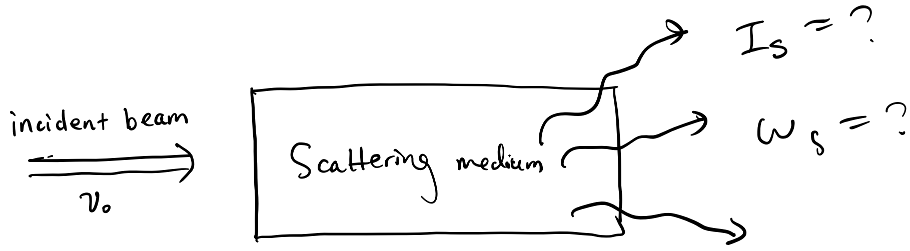
Note:

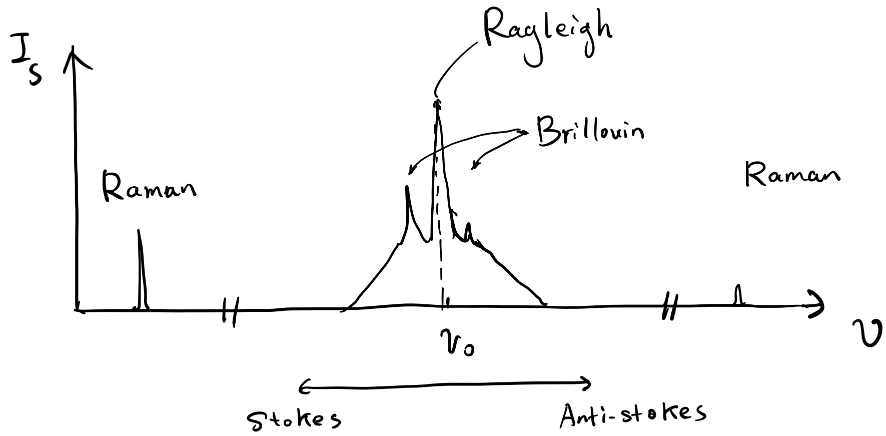
① For real-materials, there are more atoms per unit cell. ⇒ More bands

② Optical phonon ⇒ Flat band ($\Omega_{op} \sim 10^{12} \sim 10^{13}$ Hz)
Acoustic phonon ⇒ linear-like band ($\Omega_{ac} \sim 10^{10}$ Hz)

③ Monochromatic field can induce OP, but not AP.

1. Spontaneous light scattering





① **Raman scattering:**

Scattering of light by molecule vibraton (or optical phonons)

② **Brillouin scattering:**

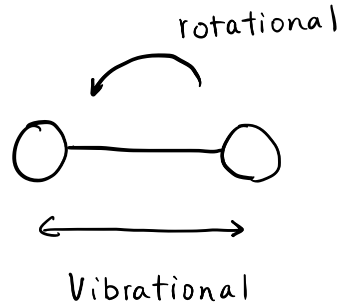
Scattering of light by sound wave (or acoustic phonon)

③ **Rayleigh-wing:**

Scattering from fluctuations in the orientation of anisotropic molecules.
(Does not occur for isotropic materials)

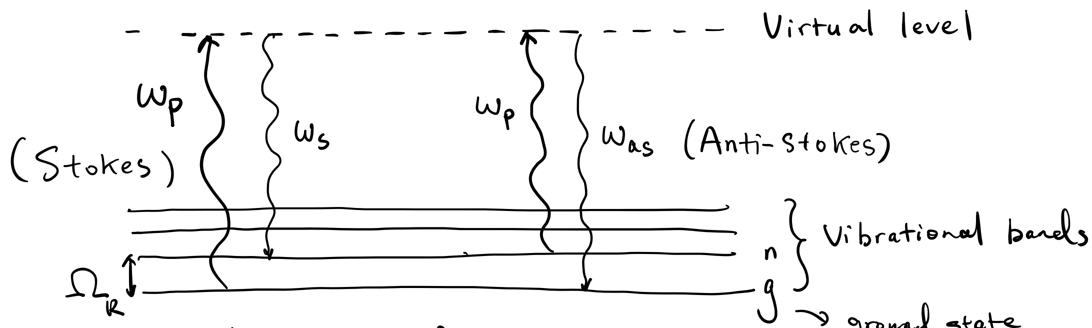
2. Spontaneous & Stimulated Raman scattering

Molecules:



local excitation ($v_s \sim 0$)

Vibration (or rotation) frequency: $\omega_v \rightarrow$ typically infrared.
($10^{12} \sim 10^{13}$ Hz)



ω_p: pump frequency.

Comments:

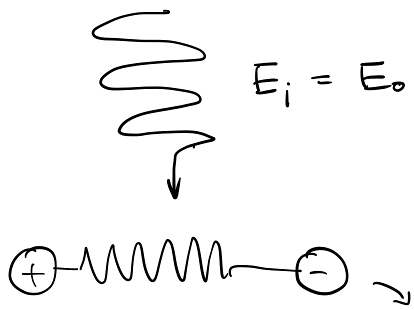
① Final state ≠ ground state (Non-parametric)

② "Stokes Raman scattering": $\omega_s < \omega_p$,
ground state vibration absorbs energy from pump, changes its energy level.

③ "Anti-Stokes Raman scattering": $\omega_s > \omega_p$. Molecules are alredy in excited state.

④ Anti-Stokes scattering is typically much weaker than Stokes. (population of n is less than g by $\exp\left(-\frac{\hbar\omega_{ng}}{kT}\right)$)

<u>Explanation:</u>



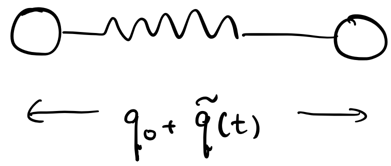
$E_i = E_0 \cos(\omega_p t)$

$p = \epsilon_0 \alpha E_i$

↳ microscopic polarizability

Key assumption: microscopic polarizability is not a constant, but depends on <u>internuclear seperation</u> $\tilde{q}(t)$.

i.e.



$$\Rightarrow \quad \alpha(q) = \alpha_0 + \left(\frac{d\alpha}{dq}\right)_0 \cdot \tilde{q}(t) + \cdots$$

$\tilde{q}(t) = q_0 \cos(\Omega_R t)$

α_0: equilibrium value

$+\cdots$: ignore, weak function of q

Microscopic polarization:

$$p \simeq \epsilon_0 \alpha \cdot E_i = \epsilon_0 \left[\alpha_0 + \left(\frac{d\alpha}{dq}\right)_0 \tilde{q}\right] E_0 \cos(\omega_p t)$$

$$= \epsilon_0 \alpha_0 E_0 \cos(\omega_p t) + \left(\frac{d\alpha}{dq}\right)_0 \tilde{q}\, E_0 \cos(\omega_p t)$$

$$= \epsilon_0 \alpha_0 E_0 \cos(\omega_p t) + \left(\frac{d\alpha}{dq}\right)_0 q_0 \underbrace{\cos(\Omega_R t) E_0 \cos(\omega_p t)}$$

$$= \epsilon_0 \alpha_0 E_0 \cos(\omega_p t) + \frac{E_0 q_0}{2}\left(\frac{d\alpha}{dq}\right)_0 \left[\cos(\Omega_R + \omega_p)t + \cos(\Omega_R - \omega_p)t\right]$$

$$= p^{(\omega_p)} + p^{(\omega_p + \Omega_R)} + p^{(\omega_p - \Omega_R)}$$

$p^{(\omega_p)}$ → Pump.

$p^{(\omega_p + \Omega_R)}$ → Anti-Stokes

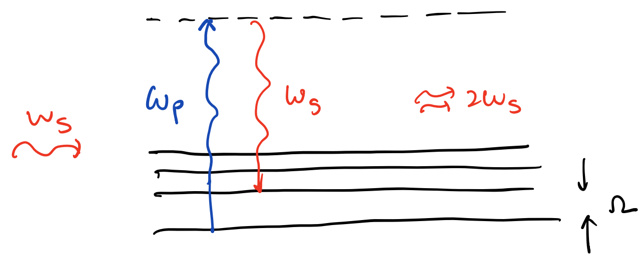
$p^{(\omega_p - \Omega_R)}$ → Stokes

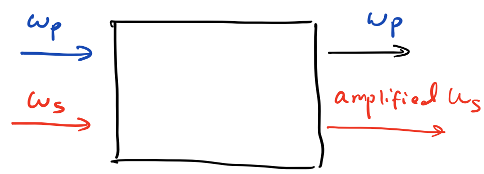
Comments:

1. Can be used to measure the vibrational levels,
   ⇒ fingerprints of various molecules (Raman spectroscopy)

2. Challenge: very weak!
   → Surface-enhanced Raman scattering (SERS)
   or Stimulated Raman.

Stimulated Raman scattering





two input lasers: one at ω_p. one at ω_s

How to detemine ω_s? Fix ω_p, scan across ω_s, monitor intensity.

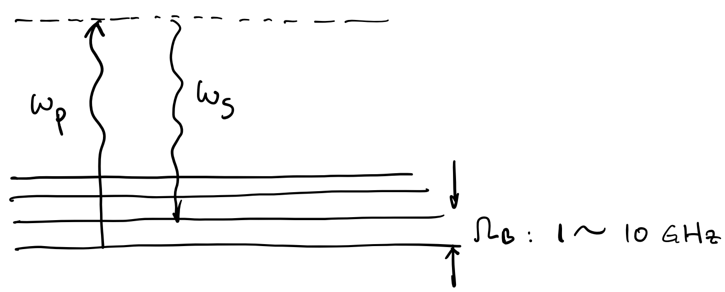
Applications: Raman amplifier, Raman laser...

3. Spontaneous & Stimulated Brillouin Scattering.

Brillouin: Acoustic wave in bulk materials (global excitation, $v_s \neq 0$)

Microwave frequencies. (1~10 GHz)

level diagram look similar to Raman

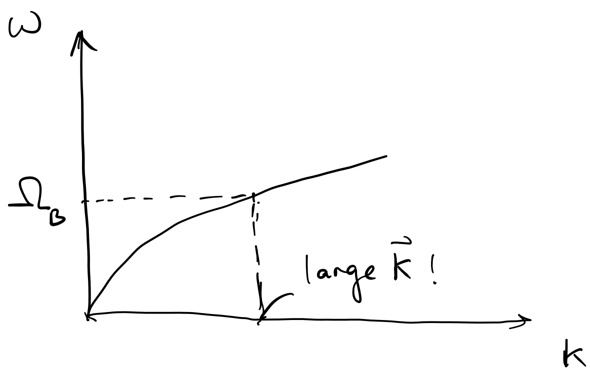


Input: optical pump at ω_p.

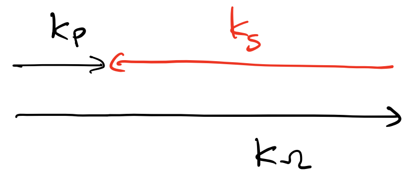
Output: optical field at ω_s, and acoustic wave at $\Omega_B = \omega_p - \omega_s$

Spontaneous Brillouin scattering

In Bulk medium:

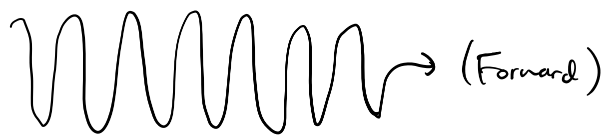


Momentum conservation

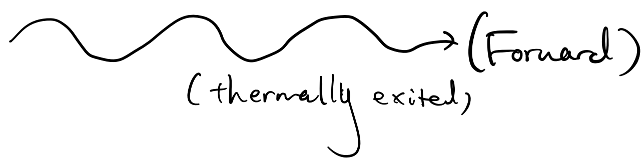


$k_\Omega > k_p$, k_s has to be negative (Backward)

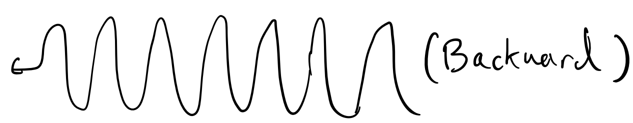
① Optical pump at ω_p.



② Output acoustic wave

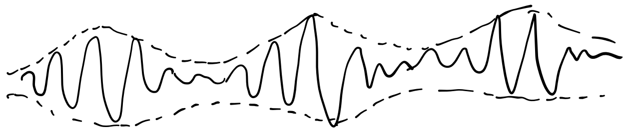


③ Output Stokes wave



Stimulated Brillouin Scattering (SBS)

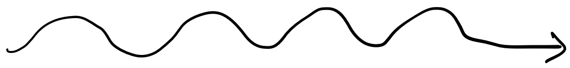
① $A_p e^{ik_p x} + A_s e^{-ik_s x}$



$\Rightarrow$ Fringes.

② Interfere fringes generate acoustic wave through "electrostriction"

↖ Materials get compressed by external field



(thermal + optically excited)

③ the generated acoustic wave act as grating, generates more reflection at ω_s