Chapter 8

Spontaneous Light Scattering and Acoustooptics

By spontaneous light scattering, we mean light scattering under conditions such that the optical properties of the material system are unmodified by the presence of the incident light beam.
(a) incident beam frequency $\nu_0$  

Scattering medium  
Scattering light  

(b) $I_s$  

Stokes  
Antistokes  

Raman  
Rayleigh  
Brillouin  
Rayleigh wing  

(Molecular orientations)  
(Entropy fluctuations)  
(Acoustic phonons)  
(Optical phonons)
<table>
<thead>
<tr>
<th>Process</th>
<th>Shift (cm(^{-1}))</th>
<th>Linewidth (cm(^{-1}))</th>
<th>Relaxation Time (sec)</th>
<th>Gain (^a) (m/MW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raman</td>
<td>1000</td>
<td>5</td>
<td>(10^{-12})</td>
<td>(5 \times 10^{-5})</td>
</tr>
<tr>
<td>Brillouin</td>
<td>0.1</td>
<td>(5 \times 10^{-3})</td>
<td>(10^{-9})</td>
<td>(10^{-4})</td>
</tr>
<tr>
<td>Rayleigh</td>
<td>0</td>
<td>(5 \times 10^{-4})</td>
<td>(10^{-8})</td>
<td>(10^{-6})</td>
</tr>
<tr>
<td>Rayleigh-wing</td>
<td>0</td>
<td>5</td>
<td>(10^{-12})</td>
<td>(10^{-5})</td>
</tr>
</tbody>
</table>

\(^a\) Gain of the stimulated version of the process.
Fluctuations as the Origin of Light Scattering

- A completely homogeneous system can scatter light only in the forward direction.
- Coherent forward scattering is the origin of refractive index.
- Small fluctuations in density in $dV_1$ and $dV_2$ can degrade the destructive interference
Fluctuations as the Origin of Light Scattering

\[ \epsilon_{ik} = \epsilon \delta_{ik} + \Delta \epsilon_{ik}, \]

\[ \Delta \epsilon_{ik} = \Delta \epsilon \delta_{ik} + \Delta \epsilon^{(t)}_{ik} \]

Scalar light scattering from fluctuations in P, S, T, \( \rho \)
Brillouin and Rayleigh scattering

Diagonal tensor
Traceless tensor
Tensor Light Scattering
Also called depolarized scattering

\[ \Delta \epsilon^{(s)}_{ik} = \Delta \epsilon^{(t)}_{ik} + \Delta \epsilon^{(a)}_{ik} \]

Symmetric
Antisymmetric
Rayleigh-wing scattering
Raman scattering

It can be shown that the fluctuations \( \Delta \epsilon \), \( \Delta \epsilon^{(s)}_{ik} \), and \( \Delta \epsilon^{(a)}_{ik} \) are statistically independent.
Scattering Cross section

\[ \frac{dP}{d\Omega} = I_0 R V \]

- \( R \): Scattering Coefficient
  - \( \sim 2 \times 10^{-8} \text{ cm}^{-1} \) for air
  - \( \sim 1.4 \times 10^{-6} \text{ cm}^{-1} \) for water

Solid angle
\[ d\Omega = \frac{dA}{L^2} \]

Power
\[ dP = I_s dA \]

Valid for dilute medium; entirely invalid for condensed matter

\[ R = \frac{\mathcal{N} \, d\sigma}{V \, d\Omega} \]

- \( \mathcal{N} \): Number of scatterers
- \( \sigma \): Scattering cross section

Differential Scattering cross section
Scattering from a Dilute Medium

Assume:
Scattering particle small compared to optical wavelength
Hence scattering only due to electric dipole and not multipole

\[ \tilde{p} = \epsilon_0 \alpha(\omega) E_0 e^{-i\omega t} + \text{c.c.} \]

\[ I_s = \frac{n\langle \tilde{p}^2 \rangle}{16\pi^2\epsilon_0 c^3 L^2} \sin^2 \phi = \frac{n\omega^4 \epsilon_0 |\alpha(\omega)|^2 |E_0|^2}{8\pi^2 c^3 L^2} \sin^2 \phi. \]

\[ \frac{d\sigma}{d\Omega} = \frac{1}{16\pi^2} \frac{\omega^4}{c^4} |\alpha(\omega)|^2 \sin^2 \phi. \]

\[ \left( \frac{d\sigma}{d\Omega} \right)_{\text{unpolarized}} = \frac{1}{32\pi^2} \frac{\omega^4}{c^4} |\alpha(\omega)|^2 (1 + \cos^2 \theta) \]
Scattering from a Lorentz Oscillator

\[ \sigma = \frac{8\pi}{3} \left( \frac{e^2}{4\pi \varepsilon_0 mc^2} \right)^2 \frac{\omega^4}{(\omega_0^2 - \omega^2)^2 + 4\omega^2 \gamma^2} \]

\[ \alpha(\omega) = \frac{e^2/Me}{\omega_0^2 - \omega^2 - 2i\omega\gamma} \]

Classical electron radius
- approximately 2.82 fm

Rayleigh scattering
- for \( \omega \ll \omega_0 \)

Thompson scattering
- for \( \omega \gg \omega_0 \)

Polarizability
Scattering from Small Dielectric Particles

\[ \alpha = 4\pi \epsilon a^3 \left( \frac{\epsilon_1 - \epsilon}{\epsilon_1 + 2\epsilon} \right)^\text{volume} \]

\[ \sigma = 16\pi^2 \frac{8\pi}{3} \frac{\omega^4}{c^4} a^6 \epsilon^2 \left( \frac{\epsilon_1 - \epsilon}{\epsilon_1 + 2\epsilon} \right)^2 \left( \frac{\epsilon_1 - \epsilon}{\epsilon_1 + 2\epsilon} \right)^\text{volume^2} \]
We consider the case in which light scattering occurs as the result of fluctuations in the (scalar) dielectric constant and in which these fluctuations are themselves the result of fluctuations in thermodynamic variables, such as the material density and temperature.
Scattered intensity from one cell: \[ I_s = I_0 \frac{\omega^4 V' V' \langle \Delta \epsilon^2 \rangle \sin^2 \phi}{16 \pi^2 L^2 c^4} \]

Change in dielectric permittivity in \( V' \): \[ \Delta \epsilon = \left( \frac{\partial \epsilon}{\partial \rho} \right)_T \Delta \rho + \left( \frac{\partial \epsilon}{\partial T} \right)_\rho \Delta T \]

Mean square deviation: \[ \langle \Delta \epsilon^2 \rangle = \left( \frac{\partial \epsilon}{\partial \rho} \right)^2 \langle \Delta \rho^2 \rangle = \gamma_e^2 \frac{\langle \Delta \rho^2 \rangle}{\rho_0^2} \]

Temperature changes to density: \[ \frac{\langle \Delta \rho^2 \rangle}{\rho_0^2} = \frac{kT}{V'} \frac{C_T}{\rho_0} \]

Isothermal compressibility: \[ C_T = -\frac{1}{V} \left( \frac{\partial V}{\partial \rho} \right)_T \]

Additional dipole moment: \[ \tilde{p} = \varepsilon_0 V' \tilde{P} = \varepsilon_0 \Delta \epsilon V' \tilde{E}_0. \]

Electrostrictive constant: \[ \gamma_e = \left( \frac{\rho}{\partial \rho} \right)_{\rho=\rho_0} \]

Electrostrictive constant: \[ \frac{\omega^4 V}{16 \pi^2 L^2 c^4} \gamma_e^2 C_T kT \sin^2 \phi \]

Reflection coefficient: \[ R = \frac{\omega^4}{16 \pi^2 c^4} \gamma_e^2 C_T kT \sin^2 \phi \]
Spectrum of Scalar Light Scattering

\[ \Delta \tilde{\epsilon} = \left( \frac{\partial \epsilon}{\partial \rho} \right) \Delta \tilde{\rho}. \]

\[ \Delta \tilde{\rho} = \left( \frac{\partial \rho}{\partial p} \right)_s \Delta \tilde{p} + \left( \frac{\partial \rho}{\partial s} \right)_p \Delta \tilde{s}. \]

Tilde indicates time dependence

Brillouin scattering due to adiabatic density fluctuations
Rayleigh-center scattering due to isobaric density fluctuations

Figure 8.3.3 Spectrum showing Brillouin and Rayleigh scattering.
Brillouin Scattering

Pressure wave

\[
\frac{\partial^2 \Delta \tilde{p}}{\partial t^2} - \Gamma' \nabla^2 \frac{\partial \Delta \tilde{p}}{\partial t} - v^2 \nabla^2 \Delta \tilde{p} = 0.
\]

Sound velocity

\[
v^2 = \left( \frac{\partial p}{\partial \rho} \right)_s = \frac{K_s}{\rho} = \frac{1}{C_s \rho}
\]

Bulk modulus

Compressibility

\[
\Gamma' = \frac{1}{\rho} \left[ \frac{4}{3} \eta_s + \eta_b + \frac{\kappa}{C_p} (\gamma - 1) \right]
\]

Bulk viscosity

Thermal conductivity

Shear viscosity

Table 8.3.1 Typical sound velocities

<table>
<thead>
<tr>
<th>Material</th>
<th>( v ) (m/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gases</td>
<td></td>
</tr>
<tr>
<td>Dry air</td>
<td>( 3.31 \times 10^2 )</td>
</tr>
<tr>
<td>He</td>
<td>( 9.65 \times 10^2 )</td>
</tr>
<tr>
<td>H(_2)</td>
<td>( 12.84 \times 10^2 )</td>
</tr>
<tr>
<td>Water vapor</td>
<td>( 4.94 \times 10^2 )</td>
</tr>
<tr>
<td>Liquids</td>
<td></td>
</tr>
<tr>
<td>CS(_2)</td>
<td>( 1.15 \times 10^3 )</td>
</tr>
<tr>
<td>CCl(_4)</td>
<td>( 0.93 \times 10^3 )</td>
</tr>
<tr>
<td>Ethanol</td>
<td>( 1.21 \times 10^3 )</td>
</tr>
<tr>
<td>Water</td>
<td>( 1.50 \times 10^3 )</td>
</tr>
<tr>
<td>Solids</td>
<td></td>
</tr>
<tr>
<td>Fused silica</td>
<td>( 5.97 \times 10^3 )</td>
</tr>
<tr>
<td>Lucite</td>
<td>( 2.68 \times 10^3 )</td>
</tr>
</tbody>
</table>
Brillouin Scattering

\[ \Delta \tilde{p} = \Delta pe^{i(qz - \Omega t)} + \text{c.c.} \]

Sound wave

\[ \Omega^2 = q^2 (v^2 - i\Omega \Gamma') \]

Dispersion relation

\[ q \approx \frac{\Omega}{v} + \frac{i\Gamma}{2v} \]

Phonon decay rate

\[ \Gamma = \Gamma' q^2 \]

\[ |\Delta p(z)|^2 = |\Delta p(0)|^2 e^{-\alpha_s z} \]

Sound absorption coefficient

\[ \alpha_s = \frac{q^2 \Gamma'}{v} = \frac{\Gamma}{v} \]

\[ \nabla^2 \tilde{E} - \frac{n^2}{c^2} \frac{\partial^2 \tilde{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \tilde{P}}{\partial t^2} \]

\[ \tilde{P}(r, t) = \epsilon_0 \left( \frac{\partial \epsilon}{\partial \rho} \right) \left( \frac{\partial \rho}{\partial p} \right)_s \Delta \tilde{p}(r, t) \tilde{E}_0(z, t) \]

\[ = \epsilon_0 \gamma_e C_s \Delta \tilde{p}(r, t) \tilde{E}_0(z, t), \]

\[ \tilde{E}_0(z, t) = E_0 e^{i(k \cdot r - \omega t)} + \text{c.c} \]

Stokes scattering

\[ \nabla^2 \tilde{E} - \frac{n^2}{c^2} \frac{\partial^2 \tilde{E}}{\partial t^2} = -\frac{\gamma_e C_s}{c^2} \left[ (\omega - \Omega)^2 E_0 \Delta p^* e^{i(k - q) \cdot r - i(\omega - \Omega) t} \right. \]

\[ + (\omega + \Omega)^2 E_0 \Delta p e^{i(k + q) \cdot r - i(\omega + \Omega) t} + \text{c.c.} \]

Antistokes scattering
Brillouin Scattering

Stokes Scattering

(a) \( \mathbf{k}' = \mathbf{k} - \mathbf{q} \)
\( \theta \)

(b) \( \mathbf{k}' = \mathbf{k} - \mathbf{q} \)
\( \theta \)

(c) \( \mathbf{k}' = \mathbf{k} - \mathbf{q} \)
\( \omega' = \omega - \Omega \)
\( \mathbf{k}, \omega \)
\( \mathbf{q}, \Omega \)

Antistokes Scattering

(a) \( \mathbf{k}' = \mathbf{k} + \mathbf{q} \)
\( \theta \)

(b) \( \mathbf{k}' = \mathbf{k} + \mathbf{q} \)
\( \theta \)

(c) \( \mathbf{k}' = \mathbf{k} + \mathbf{q} \)
\( \omega' = \omega + \Omega \)
\( \mathbf{k}, \omega \)
\( \mathbf{q}, \Omega \)
Rayleigh Center Scattering

Specific heat

\[ \rho c_p \frac{\partial \Delta \tilde{s}}{\partial t} - \kappa \nabla^2 \Delta \tilde{s} = 0. \]

Entropy wave is similar to diffusion equation

\[ \Delta \tilde{s} = \Delta s_0 e^{-\delta t} e^{-i\mathbf{q} \cdot \mathbf{r}} \quad \delta = \frac{\kappa}{\rho c_p} q^2 \]

Does not propagate, so cannot shift frequency
Brillouin vs. Rayleigh-Center

\[
\frac{I_c}{2I_B} = \frac{c_p - c_v}{c_v} = \gamma - 1.
\]  

(8.3.56)

Here \( I_c \) denotes the integrated intensity of the central component, and \( I_B \) that of either of the Brillouin components. This result is known as the Landau–Placzek relation.