Lecture 10. Lidar Effective Cross-Section vs. Convolution

- Introduction
- Convolution in Lineshape Determination
  -- Voigt Lineshape (Lorentzian × Gaussian)
- Effective Cross Section for Single Isotope
  -- Na Resonance Fluorescence Lidar
- Effective Cross Section for multiple Isotopes
  -- K Resonance Fluorescence Lidar
- Convoluted Rayleigh Doppler Lineshape
  -- Influence of Laser Lineshape
Resonance fluorescence contains two single-photon processes: Absorption of a photon by an atom, and spontaneous emission of another photon by the atom. Fluorescence can be quenched by collisions.

Rayleigh scattering or Raman scattering is a two-photon process, in which two photons are involved simultaneously. Therefore, Rayleigh/Raman scattering has much smaller scattering cross-section than absorption, about $10^{14}$ & $10^{17}$ orders of magnitude smaller, respectively.

Here, 0, 1, and 2 are real energy states, while 3 is a virtual state.

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Atomic Absorption Lineshape Determined by Convolution

\[ I(\omega) = I_0 \int_{-\infty}^{+\infty} n(\omega', \omega_0) g_{\text{Lorentzian}}(\omega', \omega) d\omega' \]

\[ \frac{I_0 (\lambda/2\pi) N \lambda}{\sqrt{2\pi} (2\pi \sigma_D)} \int_{0}^{\infty} \exp \left[ -\frac{(\omega' - \omega_0)^2}{2 (2\pi \sigma_D)^2} \right] \frac{d\omega'}{(\omega - \omega')^2 + (\lambda/2)^2} \]
Natural Linewidth Determined by Finite Radiative Lifetime

Usually only the ground state can have infinite radiative lifetime, therefore infinite thin energy level. Excited states all have finite radiative lifetime, therefore finite energy level widths.
Natural Linewidth Determined by Finite Radiative Lifetime

Infinite lifetime $\rightarrow$ harmonic oscillator $\rightarrow$ narrow linewidth

Finite radiative lifetime $\rightarrow$ damped oscillator $\rightarrow$ natural linewidth

In frequency domain, the $\delta(w-w_0)$ function is replaced by a line profile with certain width and shape.
Atomic Absorption Cross-Section

- $\sigma_{\text{abs}}$ is proportional to the probability of a single-frequency photon being absorbed by an atom:

$$\sigma_{\text{abs}}(\nu, \nu_0) = A_{ki} \frac{\lambda^2}{8\pi n^2} \frac{g_k}{g_i} g_A(\nu, \nu_0)$$  \hspace{1cm} (10.1)

where $A_{ki}$ is the spontaneous transition probability per unit time, i.e., the Einstein $A$ coefficient; $g_k$ and $g_i$ are the degeneracy factors for the upper and lower energy levels $k$ and $i$, respectively; $\lambda$ is the wavelength; $n$ is the refraction index, and $g_A$ is the absorption lineshape. $\nu$ and $\nu_0$ are the laser frequency and the central frequency of the atomic absorption line, respectively.

- For single atom, the absorption lineshape $g_A$ is determined by natural linewidth and collisional broadening, which has Lorentzian shape

$$g_A(\nu, \nu_0) = g_H(\nu, \nu_0) = \frac{\Delta \nu_H}{2\pi \left[ (\nu - \nu_0)^2 + (\Delta \nu_H / 2)^2 \right]}$$  \hspace{1cm} (10.2)

where $\Delta \nu_H$ is the homogeneous broadened linewidth.

Refer to our textbook Chapter 5 and references therein.
Absorption Cross Section Under Doppler Effects

- For many atoms in thermal equilibrium, the lineshape is the integration of the Doppler broadening (Gaussian shape) with the Lorentzian natural lineshape. The absorption lineshape $g_A$ is given by Voigt integration

$$g_A(\omega) = \int_{-\infty}^{+\infty} g_{\text{Gaussian}}(\omega_0, \omega') g_{\text{Lorentzian}}(\omega, \omega') d\omega'$$
Absorption Cross Section Under Doppler Effects

When ignoring the Lorentzian natural linewidth (~10 MHz), the Doppler broadening (~1 GHz) dominates the lineshape. The statically averaged absorption cross section for each atomic transition line is then given by a Gaussian

$$\sigma_{abs}(\nu, \nu_o) = \sigma_o \exp \left( - \frac{\nu(1 - V_R/c) - \nu_o}{2\sigma_D^2} \right)$$  \hspace{1cm} (10.3)

where

$$\sigma_o = \frac{1}{\sqrt{2\pi}\sigma_D} \frac{e^2}{4\epsilon_o m_e c} f_{ik}$$  \hspace{1cm} (10.4)

and

$$\sigma_D = \sqrt{\frac{k_B T}{M\lambda_0^2}}$$  \hspace{1cm} (10.5)

- $\sigma_o$ – peak absorption cross section (m$^2$ or cm$^2$)
- $e$ – charge of electron; $m_e$ – mass of electron
- $\epsilon_o$ – electric constant; $c$ – speed of light
- $f_{ik}$ – absorption oscillator strength; $V_R$ – radial velocity
- $\sigma_D$ – Doppler broadened line width
- $k_B$ – Boltzmann constant; $T$ – temperature
- $M$ – mass of atom; $\lambda_0$ – wavelength
Resonance Fluorescence Lidar: Effective Cross Section

- Refer to the textbook Section 5.2.1.3.1 for effective cross section $\sigma_{\text{eff}}$.
- Resonance fluorescence contains two single-photon processes: Absorption of a photon by an atom, and spontaneous emission of another photon by the atom.
- Because resonance fluorescence is isotropic, the differential backscatter cross-section $d\sigma/d\Omega$ can be replaced by the total effective scattering cross-section $\sigma_{\text{eff}}$ divided by $4\pi$.
- The total effective scattering cross-section $\sigma_{\text{eff}}$ is defined as the ratio of the average photon number scattered by an individual atom (in all directions) to the total incident photon number per unit area.
- The $\sigma_{\text{eff}}$ is determined by the convolution of the atomic absorption cross-section $\sigma_{\text{abs}}$ and the laser spectral lineshape $g_L(\nu)$.
- The absorption cross-section $\sigma_{\text{abs}}$ is defined as the ratio of the average absorbed single-frequency photons per atom to the total incident photons per unit area.
Na Atomic Energy Levels

- Na (sodium) is probably one of the most well studied alkali species.
- $^{23}\text{Na}$ is the only stable isotope for Na, and it has hyperfine structures.

Refer to our textbook Chapter 5 and references therein.
# Na Atomic Parameters

## Table 5.1 Parameters of the Na D₁ and D₂ Transition Lines

<table>
<thead>
<tr>
<th>Transition Line</th>
<th>Central Wavelength (nm)</th>
<th>Transition Probability ($10^8$ s⁻¹)</th>
<th>Radiative Lifetime (nsec)</th>
<th>Oscillator Strength $f_{ik}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>D₁ ($^2P_{1/2} \rightarrow ^2S_{1/2}$)</td>
<td>589.7558</td>
<td>0.614</td>
<td>16.29</td>
<td>0.320</td>
</tr>
<tr>
<td>D₂ ($^2P_{3/2} \rightarrow ^2S_{1/2}$)</td>
<td>589.1583</td>
<td>0.616</td>
<td>16.23</td>
<td>0.641</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Group</th>
<th>$^2S_{1/2}$</th>
<th>$^2P_{3/2}$</th>
<th>Offset (GHz)</th>
<th>Relative Line Strength$^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>D₂b</td>
<td>$F = 1$</td>
<td>$F = 2$</td>
<td>1.0911</td>
<td>5/32</td>
</tr>
<tr>
<td></td>
<td>$F = 1$</td>
<td></td>
<td>1.0566</td>
<td>5/32</td>
</tr>
<tr>
<td></td>
<td>$F = 0$</td>
<td></td>
<td>1.0408</td>
<td>2/32</td>
</tr>
<tr>
<td>D₂a</td>
<td>$F = 2$</td>
<td>$F = 3$</td>
<td>−0.6216</td>
<td>14/32</td>
</tr>
<tr>
<td></td>
<td>$F = 2$</td>
<td></td>
<td>−0.6806</td>
<td>5/32</td>
</tr>
<tr>
<td></td>
<td>$F = 1$</td>
<td></td>
<td>−0.7150</td>
<td>1/32</td>
</tr>
</tbody>
</table>

## Doppler-Free Saturation–Absorption Features of the Na D₂ Line

<table>
<thead>
<tr>
<th>$f_a$ (MHz)</th>
<th>$f_c$ (MHz)</th>
<th>$f_b$ (MHz)</th>
<th>$f_+$ (MHz)</th>
<th>$f_-$ (MHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>−651.4</td>
<td>187.8</td>
<td>1067.8</td>
<td>−21.4</td>
<td>−1281.4</td>
</tr>
</tbody>
</table>

$^a$Relative line strengths are in the absence of a magnetic field or the spatial average. When Hanle effect is considered in the atmosphere, the relative line strengths will be modified depending on the geomagnetic field and the laser polarization.
Doppler-Limited Na Spectroscopy

- Doppler-broadened Na absorption cross-section is approximated as a Gaussian with rms width $\sigma_D$

$$\sigma_{abs}(\nu) = \frac{1}{\sqrt{2\pi}\sigma_D} \frac{e^2 f}{4\epsilon_0 m_e c} \sum_{n=1}^{6} A_n \exp\left(-\frac{[\nu_n - \nu(1-V_R/c)]^2}{2\sigma_D^2}\right)$$ (10.6)

- Assume the laser lineshape is a Gaussian with rms width $\sigma_L$

- The effective cross-section is the convolution of the atomic absorption cross-section and the laser lineshape

$$\sigma_{eff}(\nu) = \frac{1}{\sqrt{2\pi}\sigma_e} \frac{e^2 f}{4\epsilon_0 m_e c} \sum_{n=1}^{6} A_n \exp\left(-\frac{[\nu_n - \nu(1-V_R/c)]^2}{2\sigma_e^2}\right)$$ (10.7)

where

$$\sigma_e = \sqrt{\sigma_D^2 + \sigma_L^2}$$ (10.8) and $$\sigma_D = \sqrt{\frac{k_B T}{M\lambda_0^2}}$$ (10.5)
Effective Cross-Section

- The effective cross section is a convolution of the atomic absorption cross section and the laser line shape. When the laser has finite spectral linewidth with lineshape (its intensity distribution along frequency) of

\[
\int_{0}^{\infty} g_L(\nu, \nu_L) d\nu = 1 \quad (10.9)
\]

- The effective cross-section \(\sigma_{\text{eff}}\) is given by the convolution of \(\sigma_{\text{abs}}\) with the laser lineshape \(g_L\):

\[
\sigma_{\text{eff}}(\nu_L, \nu_o) = \int_{-\infty}^{+\infty} \sigma_{\text{abs}}(\nu, \nu_o) g_L(\nu, \nu_L) d\nu \quad (10.10)
\]

- If the laser spectral lineshape is a Gaussian shape:

\[
g_L(\nu, \nu_L) = \frac{1}{\sqrt{2\pi}\sigma_L} \exp\left(-\frac{(\nu - \nu_L)^2}{2\sigma_L^2}\right) \quad (10.11)
\]

- Then \(\sigma_{\text{eff}}\) can be written as

\[
\sigma_{\text{eff}}(\nu_L, \nu_0) = \frac{\sigma_D \sigma_0}{\sqrt{\sigma_D^2 + \sigma_L^2}} \exp\left(-\frac{[\nu_L (1 - V_R/c) - \nu_0]^2}{2(\sigma_D^2 + \sigma_L^2)}\right) \quad (10.12)
\]
Effective Cross-Section for K Atoms

Atomic K energy levels
Reference our textbook Chapter 5

K (potassium) is another alkali species, and it has several stable isotopes (\(^{39}\)K, \(^{40}\)K, and \(^{41}\)K) with hyperfine structures.
Effective Cross-Section for K Atoms

Absorption cross section of K atom’s D1 line is given by

\[
\sigma_{\text{abs}}(\nu) = \sum_{A=39}^{41} \left\{ \text{Isotope Abdn}(A) \frac{1}{\sqrt{2\pi}\sigma_D} \frac{e^2 f}{4\varepsilon_0 m_e c} \sum_{n=1}^{4} A_n \exp \left( -\frac{[\nu_n - \nu(1 - V_R/c)]^2}{2\sigma_D^2} \right) \right\}
\]

Isotope abundance: 93.2581% (\textsuperscript{39}K), 0.0117% (\textsuperscript{40}K), 6.7302% (\textsuperscript{41}K)

Line strength: \( A_n = \frac{5}{16}, \frac{1}{16}, \frac{5}{16}, \frac{5}{16} \)

Oscillator strength: \( f \), Doppler broadening: \( \sigma_D \)

The effective total scattering cross section of K atom’s D1 line is the convolution of the absorption cross section and the laser lineshape. Under the assumption of Gaussian lineshape of the laser, it is given by

\[
\sigma_{\text{eff}}(\nu) = \sum_{A=39}^{41} \left\{ \text{Isotope Abdn}(A) \frac{1}{\sqrt{2\pi}\sigma_e} \frac{e^2 f}{4\varepsilon_0 m_e c} \sum_{n=1}^{4} A_n \exp \left( -\frac{[\nu_n - \nu(1 - V_R/c)]^2}{2\sigma_e^2} \right) \right\}
\]

where

\[
\sigma_e = \sqrt{\sigma_D^2 + \sigma_L^2} \quad (11.11)
\]

and

\[
\sigma_D = \sqrt{\frac{k_B T}{M\lambda_0^2}} \quad (11.12)
\]

Refer to our textbook Chapter 5 and references therein.
Convoluted Rayleigh Doppler Lineshape

If scattering is produced with a single-frequency laser beam and without pressure broadening, Rayleigh scattering from atmosphere molecules experiences pure Doppler effects (Doppler shift and Doppler broadening) and exhibits a Gaussian shape.

When the laser has finite spectral linewidth with lineshape, the return Rayleigh Doppler lineshape is a convolution of the pure Doppler Gaussian with the laser line shape.

\[
\sigma_{\text{eff}}(\nu_L, \nu_o) = \int_{-\infty}^{+\infty} \sigma_{\text{abs}}(\nu, \nu_o) g_L(\nu, \nu_L) d\nu
\]

where the laser spectral lineshape is normalized:

\[
\int_{0}^{\infty} g_L(\nu, \nu_L) d\nu = 1
\]

If the laser spectral lineshape is a Gaussian,

\[
g_L(\nu, \nu_L) = \frac{1}{\sqrt{2\pi}\sigma_L} \exp\left(-\frac{(\nu - \nu_L)^2}{2\sigma_L^2}\right)
\]

then the convolution leads to another Gaussian with RMS linewidth

\[
\sigma_{\text{convoluted}} = \sqrt{\sigma_D^2 + \sigma_L^2}
\]