Rotational Raman Scattering

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What is Raman scattering?

When light is scattered from an atom or molecule, most photons are elastically scattered (Rayleigh scattering). The scattered photons have the same energy (frequency) or wavelength as the incident photons. However, a small fraction of the scattered light is scattered with the scattered photons having a frequency different from the frequency of the incident photons (lower frequency: Stokes lines; higher frequency: anti-Stokes lines. In a gas, Raman scattering can occur with a change in vibrational, rotational or electronic energy of a molecule. Chemists are concerned primarily with the vibrational Raman effect.[1]

Rotational Raman scattering is important in the Earth's atmosphere and leads to filling in of line spectra (Ring effect).

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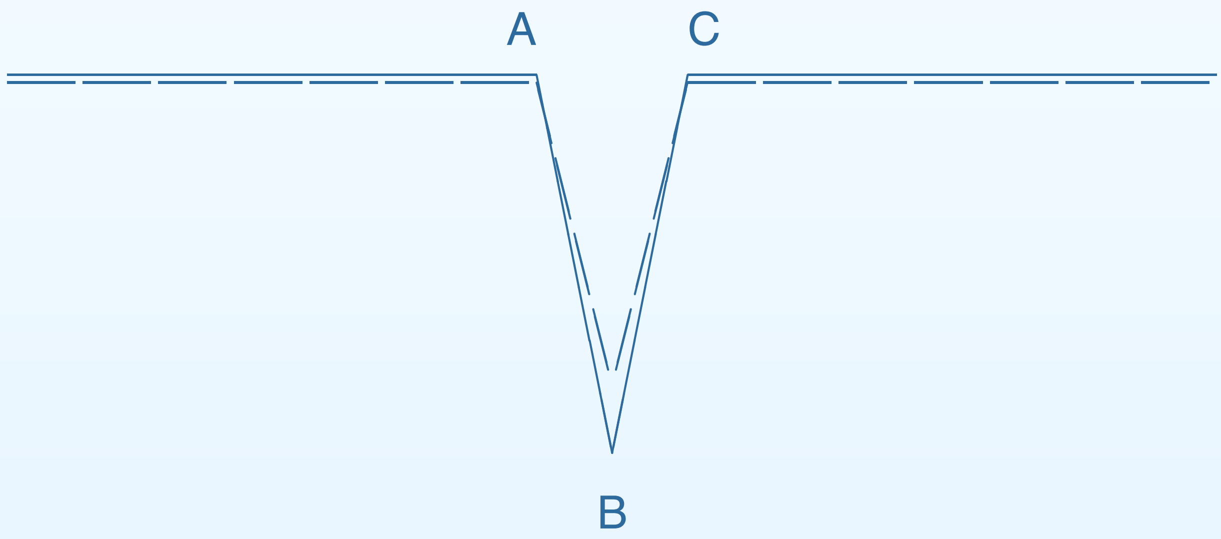
[1] Adopted wikipedia.

A naive example of filling in of line spectra

	A	B	C
Initial value	100	50	100
loss (scattered out of wavelength)	20	10	20
gain (scattered into from lower wavelength)	10	10	5
gain (scattered into from higher wavelength)	5	10	10
Total after scattering	95	60	95



The RTE including Raman scattering

Energy conservation requires[2]

$$\mathrm{d}E = \mathrm{d}I(z, \phi, \theta, \lambda_k)\mathrm{d}\Omega\mathrm{d}\lambda = \mathrm{d}E_{emi} - \mathrm{d}E_{att}$$

where, excluding scattering of aerosols and clouds for simplicity,

$$\begin{aligned} E_{att} &= \beta_{ext}(z, \lambda_k) I(z, \phi, \theta, \lambda_k)\mathrm{d}\Omega\mathrm{d}\lambda \\ E_{emi} &= \beta_{ray}(z, \lambda_k) \int \frac{\mathrm{d}\Omega'}{4\pi} I(z, \Omega', \lambda_k) P_{ray}(\Omega, \Omega')\mathrm{d}\Omega\mathrm{d}\lambda \end{aligned}$$

[2] Adopted from Vountas et al., Ring effect: impact of rotational Raman scattering on radiative transfer in Earth's atmosphere, *J. Quant. Spectrosc. Radiat. Transfer*, **60 (6)**, 943-961, 1998.

The RTE including Raman scattering cont'd

The emitted energy into the volume $E_{emi} = E_{unshift} + E_{shift}$ has contributions:

$$E_{unshift} = \int \frac{\mathrm{d}\Omega'}{4\pi} I(z,\Omega',\lambda_k)[\beta_{ray}(z,\lambda_k)P_{ray}(\Omega,\Omega') - \sum_{i=1}^{L} r(z,\lambda_k,\lambda_i)P_{rrs}(\Omega,\Omega')]\mathrm{d}\Omega\mathrm{d}\lambda$$

$$E_{shift} = \sum_{i=j}^{L} r(z,\lambda_j,\lambda_k)\int \frac{\mathrm{d}\Omega'}{4\pi} I(z,\Omega',\lambda_k)P_{rrs}(\Omega,\Omega')\mathrm{d}\Omega\mathrm{d}\lambda$$

The RTE including Raman scattering cont'd

The RTE with Raman scattering may thus be written as

$$\mathcal{R}I(z, \phi, \theta, \lambda_k) = \varepsilon_{rss}(z, \phi, \theta, \lambda_k)$$

where the operator $\mathcal{R}$

$$\mathcal{R} \equiv \mu \frac{\mathrm{d}}{\mathrm{d}z} + \beta_{ext}(z, \lambda_k) - \beta_{ray}(z, \lambda_k) \int \frac{\mathrm{d}\Omega'}{4\pi} P_{ray}(z, \lambda_k, \Omega, \Omega')\mathrm{d}\Omega$$

and

$$\begin{aligned} \varepsilon_{rss}(z, \phi, \theta, \lambda_k) &= \sum_{i=j}^{L} r(z, \lambda_j, \lambda_k) \int \frac{\mathrm{d}\Omega'}{4\pi} I(z, \Omega', \lambda_j) P_{rrs}(\Omega, \Omega')\mathrm{d}\Omega \\ &\quad - \sum_{i=1}^{L} r(z, \lambda_k, \lambda_i) \int \frac{\mathrm{d}\Omega'}{4\pi} I(z, \Omega', \lambda_k) P_{rrs}(\Omega, \Omega')\mathrm{d}\Omega \end{aligned}$$

Solution of RTE

Decompose radiation field into direct and diffuse components. The direct component is the same for elastically and inelastically scattering atmospheres. Diffuse component obeys:

$$\mathcal{R}I(z,\phi,\theta,\lambda_k) = Q_{rrs}(z,\phi,\theta,\lambda_k) + Q_{el}(z,\phi,\theta,\lambda_k) + \varepsilon_{rss}(z,\phi,\theta,\lambda_k)$$

where

$$\begin{aligned}
Q_{el}(z,\phi,\theta,\lambda_k) &= \frac{F_k(z)}{4}\beta(z,\lambda_k)P(\mu,\phi,-\mu_0,\phi_0) \\
&\quad + A\mu_0\beta(z,\lambda_k)\int\frac{d\Omega'}{4\pi}P(\Omega,\Omega')E_k(z,\mu_0,\mu') \\
Q_{rrs}(z,\phi,\theta,\lambda_k) &= \frac{1}{4}\left[\sum_{j=1}^{L}F_j(z)r(z,\lambda_j,\lambda_k) - F_k(z)r(z,\lambda_k)\right]P_{rrs}(\Omega,-\mu_0,\phi_0) \\
&\quad + A\mu_0\int\frac{d\Omega'}{4\pi}\left[\sum_{j=1}^{L}r(z,\lambda_k,\lambda_j)E_j(z,\mu_0,\mu') - r(z,\lambda_k)E_k(z,\mu_0,\mu')\right]P_{rrs}(\Omega,\Omega') \\
E_j(z,\mu_0,\mu) &= F(\lambda_j)e^{-\tau_0(\lambda_j)/\mu_0}e^{-(\tau_0(\lambda_j)/\mu_0-\tau(z,\lambda_j))/\mu'} \\
F_j(z) &= F(\lambda)e^{-(\tau(z,\lambda_j)/\mu_0} \\
r(z,\lambda_k) &= \sum_{i=1}^{L}r(z,\lambda_k,\lambda_i)
\end{aligned}$$

First order solution

Formal solution:

$$I(z,\phi,\theta,\lambda_k) = \mathcal{R}^{-1}[Q_{el}(z,\phi,\theta,\lambda_k) + Q_{rrs}(z,\phi,\theta,\lambda_k) + \varepsilon_{rss}(z,\phi,\theta,\lambda_k)] \quad (1)$$

Iterative solution, set $\varepsilon_{rss} = 0$ for first step:

$$\begin{aligned} I^{(0)}(z,\phi,\theta,\lambda_k) &= I_{el}(z,\phi,\theta,\lambda_k) + \mathcal{R}^{-1}Q_{rrs}(z,\phi,\theta,\lambda_k) \\ I^{(1)}(z,\phi,\theta,\lambda_k) &= I^{(0)}(z,\phi,\theta,\lambda_k) + \mathcal{R}^{-1}\varepsilon_{rss}^{(0)}(z,\phi,\theta,\lambda_k) \end{aligned}$$