

Fundamentals of filtered Rayleigh scattering for particle-laden flows

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ABSTRACT

Laser-based gas-flow diagnostics in particle-laden flows are strongly affected by particulate Mie scattering, making conventional Rayleigh scattering impractical in many regimes. Filtered Rayleigh scattering (FRS) offers a route to recover molecular information by spectrally suppressing high-intensity particulate contributions. This work develops a general numerical optical-chain model to quantify when FRS becomes feasible in such environments, motivated by employing FRS thermometry in iron-powder flames. The model resolves and combines the laser lineshape, optional Fabry–Pérot etalon pre-filtering, vapour absorption filtering, particulate Lorenz-Mie scattering, and molecular Rayleigh scattering, and reduces the transmitted spectra to a detector-integrated signal ratio $S_g^{\text{Ray}} / S_s^{\text{Mie}}$ as a practical design metric. Example calculations show that the feasibility boundary depends strongly on particle radius and volume fraction due to the residual broadband laser-pedestal light after absorption filtering. Improving spectral purity by means of an etalon upstream of the probe volume increases the signal ratio by more than an order of magnitude for representative settings, and thus identifies regimes of particle loading and size where FRS diagnostics in particle-laden flows are practically achievable.

1. Introduction

Filtered Rayleigh scattering (FRS) is an optical diagnostic technique for gas-flow measurements, including temperature and velocity, in which molecular scattering is spectrally separated from spurious scattering by particles or solid surfaces in the field of view. The key idea behind FRS is that molecular scattering is Doppler-broadened by the molecular velocity distribution, whereas scattering from solid or liquid particulates and surfaces is effectively elastic and remains spectrally close to the incident laser frequency. In its usual implementation, the scattered light is not spectrally resolved, but filtered by an ultra-narrow-band vapour filter that suppresses the narrowband contribution while transmitting the wings of the broadened component of the scattered light. In particle-laden or seeded flows, however, this separation becomes more difficult because particulate scattering can be orders of magnitude stronger than the molecular signal. Although FRS

has been used successfully in gas-flow diagnostics (Miles et al., 1992; Boguszeko & Elliott, 2005a; Ground et al., 2023), its feasibility range in particle-laden flows remains difficult to assess without a quantitative comparison between particle-size-dependent Mie scattering and molecular Rayleigh scattering.

The present work addresses this problem with a numerical optical-chain model that follows the light from the incident laser beam, through optional Fabry–Pérot etalon pre-filtering, scattering by gas molecules and particulates, vapour absorption filtering, and spectral integration at the detector. In particular, we consider an injection-seeded frequency-doubled Nd:YAG laser with nominal $\lambda_0 = 532 \text{ nm}$ as a light source and a molecular iodine (I_2) absorption cell as a spectral filter. The example calculations are motivated by planned FRS thermometry in micron-sized iron-powder flames. Spherical iron particles are therefore used as the representative particles, while the model itself is not limited to this application.

Our aim is to investigate the combinations of particle size, particle loading, laser spectral purity, iodine filtering, and gas temperature for which filtered Rayleigh scattering becomes feasible in a particle-laden flow. The resulting ratio between the filtered molecular Rayleigh signal and the residual particulate Mie signal is used as an order-of-magnitude design metric for selecting operating conditions and identifying when additional spectral purification is required. Throughout this paper, scattering from spherical particulates is referred to as Mie scattering, while scattering from gas molecules is referred to as Rayleigh scattering. The distinction is important because the Rayleigh limit of particulate Mie scattering and molecular Rayleigh scattering share similar wavelength scaling, but describe different scatterers and enter the model through different physical quantities.

The assumptions and formulations of the optical-chain model are described in Section 2, where they are applied to the iron-particle FRS thermometry case. Section 3 summarises the resulting feasibility limits and implications for future measurements.

2. Model description

The filtered Rayleigh scattering technique is modelled as a sequential optical chain, depicted schematically in Figure 1, in which each component shapes the signal that is eventually registered by the detector. A narrow-linewidth laser light sheet, centred at frequency $\tilde{\nu}_0$ within the tunable range of the second harmonic of a seeded Nd:YAG ($\tilde{\nu}_0 = 18787.8 \text{ cm}^{-1}$ at the iodine absorption line primarily used throughout this work), illuminates an interrogation volume containing both molecular and particulate scatterers. Light scattered at angle θ into solid angle $\Delta\Omega$ passes through an iodine vapour absorption filter of length L_{cell} , maintained at temperature T_{cell} and pressure p_{cell} , before reaching the detector. As particulate Mie scattering follows the narrow incident laser spectrum, it can be selectively attenuated by an iodine absorption line, while the molecular Rayleigh

scattering spectrum is broadened and partly transmitted. The ratio of filtered molecular Rayleigh and residual particulate Mie scattering quantifies the relative contributions of the two scattering mechanisms, provided single-scattering conditions are met. The following subsections describe the mathematical formulations and assumptions for each component in the optical chain.

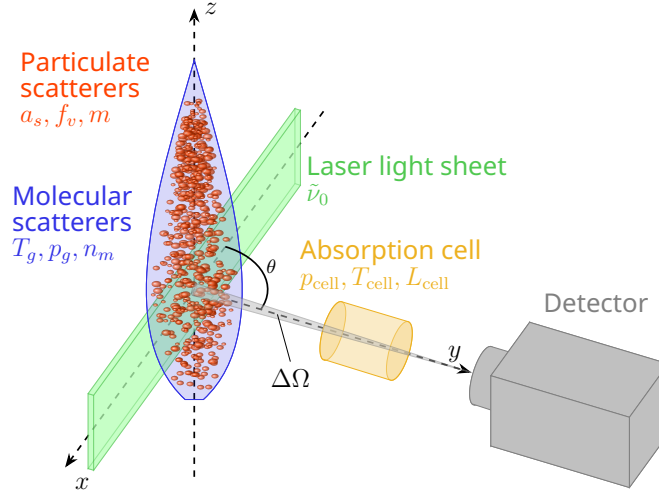


Figure 1. Schematic of the optical chain modelled in this work. Particulate scatterers (red) are attributed a sphere radius a_s , volume fraction f_v and complex refractive index m . Molecular scatterers (blue) are governed by gas temperature T_g and pressure p_g , and real refractive index n_m .

2.1. Laser spectral profile

Throughout this work, spectral positions are reported as wavenumbers because the iodine absorption spectrum is tabulated and plotted in cm^{-1} (Forkey et al., 1997). The symbol $\tilde{\nu}$ denotes the wavenumber in cm^{-1} , while $\nu = 100 \tilde{\nu}$ denotes the same wavenumber in m^{-1} . Frequency in hertz is related through the speed of light c , denoted by $f = c\nu$. Parameters are thus also specified in frequency units, unless a tilde is used to indicate the corresponding wavenumber width.

The spectral output of an injection-seeded Nd:YAG laser is modelled as a narrow single-mode Gaussian component superimposed on a broadband Lorentzian pedestal arising from amplified spontaneous emission in the gain medium (Seasholtz & Buggele, 1997; Wu et al., 2000; Patton & Sutton, 2013b). The laser spectral irradiance is therefore:

$$I_0(\tilde{\nu}) = \alpha_w I_L(\tilde{\nu}) + I_G(\tilde{\nu}), \quad (1)$$

where $I_0(\tilde{\nu})$ has units of W m^{-2} per cm^{-1} and α_w is a weighting factor. Both components are centred at $\tilde{\nu}_0$ and normalised to unit area because any common irradiance scale cancels in the ratios considered later. The Lorentzian component, with full width at half maximum (FWHM) $\tilde{\Gamma}_L$

in wavenumber units, is:

$$I_L(\tilde{\nu}) = \frac{1}{\pi} \frac{\tilde{\Gamma}_L/2}{(\tilde{\nu} - \tilde{\nu}_0)^2 + (\tilde{\Gamma}_L/2)^2}, \quad \int I_L(\tilde{\nu}) d\tilde{\nu} = 1, \quad (2)$$

and the Gaussian component, with FWHM $\tilde{\Gamma}_G$, is:

$$I_G(\tilde{\nu}) = \frac{1}{\tilde{\delta}_G \sqrt{2\pi}} \exp \left\{ -\frac{(\tilde{\nu} - \tilde{\nu}_0)^2}{2\tilde{\delta}_G^2} \right\}, \quad \int I_G(\tilde{\nu}) d\tilde{\nu} = 1, \quad (3)$$

where $\tilde{\delta}_G = \tilde{\Gamma}_G / (2\sqrt{2 \ln 2})$.

A practical figure of merit is the spectral purity (SP), which quantifies the fraction of the total laser output contained in the narrowband single-mode component (Barnes & Barnes, 1993; Lee & Lempert, 2003):

$$\text{SP} = \frac{\int I_G(\tilde{\nu}) d\tilde{\nu}}{\int I_0(\tilde{\nu}) d\tilde{\nu}} = \frac{1}{\alpha_w + 1}. \quad (4)$$

For FRS, a high spectral purity is desirable because the broadband Lorentzian pedestal is not fully attenuated by the iodine filter and is therefore the dominant residual contribution to the Mie scattering signal if the narrowband single-mode component is sufficiently absorbed. Using parameters representative of a frequency-doubled Nd:YAG laser (Sutton & Patton, 2014), $\alpha_w = 3 \cdot 10^{-5}$, $\Gamma_L = 42$ GHz, and $\Gamma_G = 75$ MHz, the resulting lineshape is shown in Figure 2. The numerical frequency grid is chosen to balance spectral accuracy and computational cost. The step size must be small enough to resolve the narrow Gaussian component, for which a spacing of approximately 30 MHz is used. The simulation window must also be wide enough to retain the broadband Lorentzian pedestal; retaining 95% of its integrated energy requires a bandwidth of $\Delta f_{\text{win}} = \Gamma_L \tan(0.95\pi/2) \approx 530$ GHz. The figures, however, show a narrower range than simulated, using the wavenumber offset from the selected absorption line on the lower axis and the corresponding frequency offset indicated on the upper axis.

2.1.1. Fabry-Pérot etalon

The broadband pedestal can be partially suppressed by passing the laser light through a Fabry-Pérot etalon (FPE) prior to the scattering volume (Seasholtz & Buggele, 1997; Patton & Sutton, 2013a; Sutton & Patton, 2014). The etalon transmission is described by the Airy function, neglecting surface absorption losses:

$$\tau_{\text{FPE}}(\tilde{\nu}) = \frac{1}{1 + \mathcal{F} \sin^2(\delta_{\text{FPE}}(\tilde{\nu})/2)}, \quad (5)$$

where $\mathcal{F} = 4\mathcal{R}/(1 - \mathcal{R})^2$ is the coefficient of finesse, $\mathcal{R}$ is the reflectivity, and the phase difference between two successively transmitted waves is:

$$\delta_{\text{FPE}}(\tilde{\nu}) = 4\pi n_{\text{FPE}} d_{\text{FPE}} (100\tilde{\nu}) \cos(\theta_{\text{FPE}}). \quad (6)$$

Here, n_{FPE} is the refractive index of the etalon medium, d_{FPE} is the etalon spacing, θ_{FPE} is the angle of incidence, and the factor 100 converts the wavenumber from cm^{-1} to m^{-1} . The spacing is chosen as $d_{\text{FPE}} = q_{\text{FPE}} / (2 n_{\text{FPE}} \cos(\theta_{\text{FPE}}) \nu_0)$, where $q_{\text{FPE}} \in \mathbb{N}$ is the interference order such that a transmission peak is centred at $\tilde{\nu}_0$, equivalently at $\nu_0 = 100\tilde{\nu}_0$. The FWHM of a transmission peak follows analytically as $\Gamma_{\text{FPE}} = c / (2 \mathcal{F} n_{\text{FPE}} d_{\text{FPE}})$, where $\mathcal{F} = (\pi/2)\sqrt{\mathcal{F}}$ is the finesse (Hecht, 2017). For the etalon to suppress the pedestal without clipping the single-mode peak, Γ_{FPE} must be greater than Γ_G . For an air-spaced etalon with $\mathcal{F} = 24.6$ and $d_{\text{FPE}} \approx 5$ mm (Sutton & Patton, 2014), the transmission profile is shown in Figure 3, giving $\Gamma_{\text{FPE}} = 1.2$ GHz. The spectral purity is:

$$\text{SP}_{\text{FPE}} = \frac{\int I_G(\tilde{\nu}) \cdot \tau_{\text{FPE}}(\tilde{\nu}) d\tilde{\nu}}{\int I_0(\tilde{\nu}) \cdot \tau_{\text{FPE}}(\tilde{\nu}) d\tilde{\nu}}, \quad (7)$$

which is evaluated numerically on the common spectral grid. The improved laser light lineshape and spectral purity after etalon filtering are included in Figure 2. The laser light incident on the probe volume is thus defined as:

$$I_i(\tilde{\nu}) = \begin{cases} I_0(\tilde{\nu}), & \text{without FPE,} \\ I_0(\tilde{\nu}) \tau_{\text{FPE}}(\tilde{\nu}), & \text{with FPE.} \end{cases} \quad (8)$$

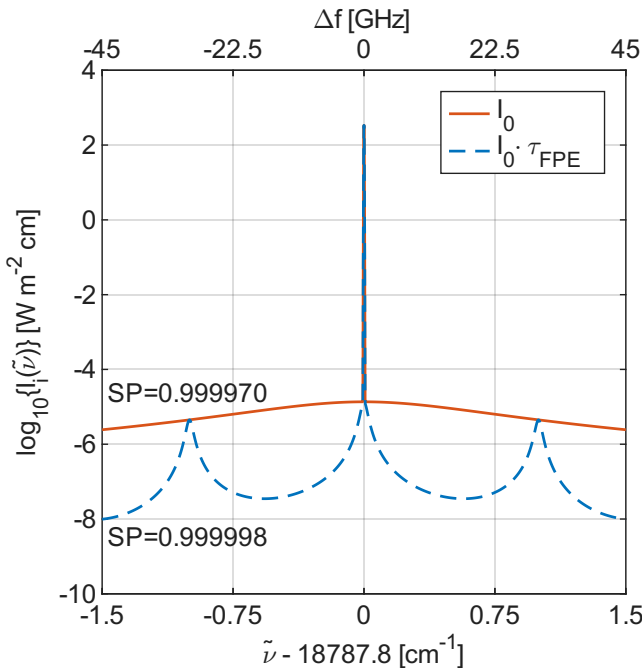


Figure 2. Spectral irradiance of the laser (log scale) before and after Fabry-Pérot etalon filtering, centred at $\tilde{\nu}_0$. Spectral purities are indicated for both profiles.

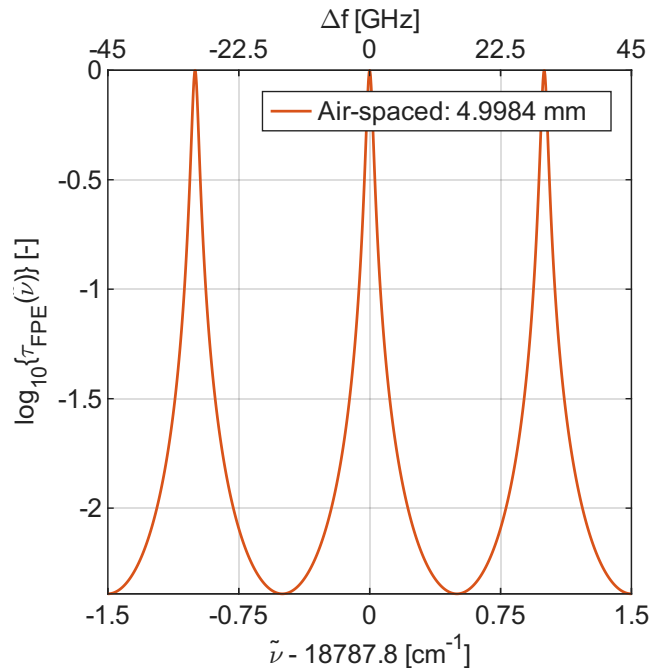


Figure 3. Fabry-Pérot transmission (log scale) modelled by the Airy function, Equation (5), for an air-spaced etalon, with a transmission peak centred at $\tilde{\nu}_0$.

2.2. Absorption filter

The choice of absorption filter material is closely linked to the laser wavelength, as ideally an optically dense and spectrally isolated absorption line aligns with the incident wavelength. A shorter laser wavelength is generally desirable due to the λ^{-4} dependence of Rayleigh scattering efficiency, but this typically comes at the cost of reduced laser power and a weaker signal. The present model uses a passive iodine vapour filter matched to the second harmonic of a Nd:YAG laser, following the established frequency-doubled Nd:YAG–I₂ configuration used in earlier FRS studies (Forkey et al., 1996; Elliott et al., 2001; Boguszko & Elliott, 2005a,b; Sutton & Patton, 2014). In a passive filter, the absorbing species directly attenuates the scattered light without requiring optical pumping of the filter medium. The iodine transmission spectrum is computed with the Forkey model as a function of side arm temperature, saturation pressure, and cell length (Forkey et al., 1997), as shown in Figure 4.

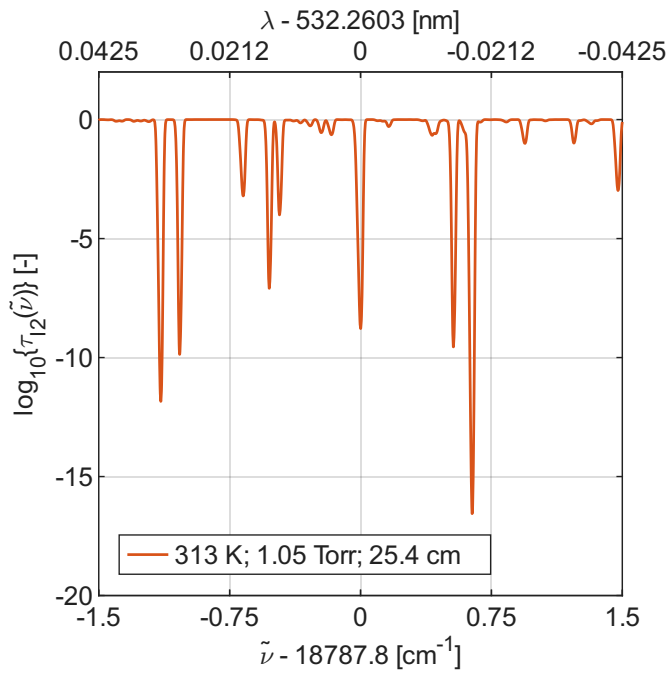


Figure 4. Iodine transmission spectrum τ_{I_2} (log scale) computed from the Forkey model (Forkey et al., 1997). The side arm temperature, saturation pressure, and cell length are aligned with those used in Sutton & Patton (2014).

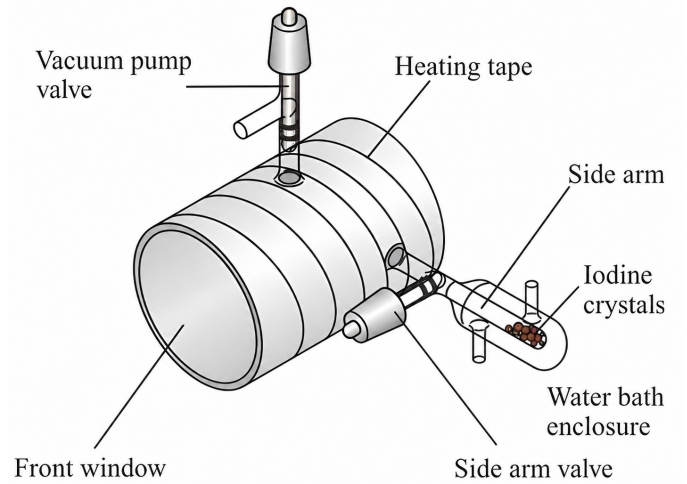


Figure 5. Schematic of the iodine vapour absorption cell. The sidearm temperature sets the iodine vapour pressure in a starved cell configuration. Maintaining an increased cell body temperature $T_{\text{cell}} > T_{\text{stem}}$ prevents condensation. Adapted from Boguszko & Elliott (2005a).

A schematic of a typical filter design is shown in Figure 5. The iodine number density inside the cell is regulated by the side arm temperature. With the side arm valve open, the vapour pressure settles at the saturation pressure at the side arm temperature. Once equilibrium is reached, the side arm is sealed, leaving the cell at a fixed number density since no additional iodine is available to increase

vapour pressure; this configuration is known as a starved cell. The cell is maintained well above the side arm temperature by means of heating tape, such that the vapour density is determined by the coldest point and condensation in the cell body is prevented (Boguszko & Elliott, 2005a,b).

2.3. Light scattering from matter

2.3.1. Scattering assumptions and terminology

The scattered light intensity is modelled under single-scattering conditions. Contributions from different scatterers in the probe volume are added as intensities rather than as coherent field amplitudes. This is appropriate when the scatterers are randomly distributed, their individual positions are unresolved by the collection optics, and the optical mean free path for particle scattering is large compared with the illuminated probe volume. Spectral broadening of the molecular contribution is treated separately from the scattering strength, because it redistributes the Rayleigh-scattered light in frequency, described later by I_{RBS} , but does not change the differential scattering cross section.

Under these conditions, the scattered spectral radiant intensity in $\text{W sr}^{-1} \text{ per cm}^{-1}$ from a probe volume V_p and number density N_j is:

$$\frac{dP_j}{d\Omega} = \frac{dC_j}{d\Omega} N_j V_p I_j(\tilde{\nu}), \quad (9)$$

where $j = s$ denotes particulate scattering and $j = g$ denotes molecular gas scattering. The spectral irradiance leaving the probe volume is therefore distinguished through:

$$I_j(\tilde{\nu}) = \begin{cases} I_i(\tilde{\nu}), & \text{Mie scattering,} \\ I_i(\tilde{\nu}) * I_{\text{RBS}}(\tilde{\nu}), & \text{Rayleigh scattering,} \end{cases} \quad (10)$$

with the asterisk indicating the convolution between the incident and broadened lineshapes. The differential scattering cross section $dC_j/d\Omega$ describes the scattered power per unit solid angle per unit incident irradiance. This angular-differential quantity is used throughout because the detector collects light over only a finite (and relatively small) solid angle. It is convenient to define the volume scattering function:

$$\beta_j = \frac{1}{V \cdot I_j(\tilde{\nu})} \frac{dP_j}{d\Omega} = \frac{dC_j}{d\Omega} N_j. \quad (11)$$

The central task remains to establish $dC_j/d\Omega$ for the particulate and molecular components. Here, a distinction must be made between Mie scattering of spheres of arbitrary size evaluated with the Lorenz-Mie solution, and Mie scattering in the Rayleigh limit, which refers to the small-size-parameter approximation indicated by $\chi_s = 2\pi a_s/\lambda_0 \ll 1$ and $\chi_s|m| \ll 1$. The term Rayleigh

scattering therefore remains reserved for molecular scattering. Although the small-sphere limit and molecular Rayleigh scattering both show a λ^{-4} -type dependence, they describe different scatterers and should not be confused.

2.3.2. Differential scattering cross sections

For scatterers whose size is small compared with the wavelength, only the lowest induced multipoles, typically the electric and possibly magnetic dipoles, contribute to the far-field radiation (Jackson, 1999). Here, the electric dipole moment $\mathbf{p}$ is of particular interest. The induced dipole moment is determined by the (scalar) polarisability α of the scatterer through:

$$\mathbf{p} = \varepsilon_0 \alpha \mathbf{E}_i, \quad (12)$$

with ε_0 the vacuum electric permittivity and $\mathbf{E}_i$ the incident electric field. The simplest case is a monochromatic plane wave incident on a small dielectric sphere of radius a_s . There is no magnetic dipole contribution for the non-conducting sphere, and the electric dipole moment at optical wavelengths is expressed as (Jackson, 1999):

$$\mathbf{p} = 4\pi\varepsilon_0 \frac{m^2 - 1}{m^2 + 2} a_s^3 \mathbf{E}_i. \quad (13)$$

The relative complex index of refraction m is defined as $m = m_s/n_m$, where n_m is the refractive index of the non-absorbing surrounding medium and $m_s = n_s + i\kappa_s$ the complex refractive index of the particle in free space, with n_s and κ_s the refractive and extinction indices of the particle, respectively. Inserting $\mathbf{p}$ into the far-field radiation formulation yields the Rayleigh-limit sphere differential cross section, which also depends on the polarisation direction of the incident light relative to the scattering plane (Jackson, 1999):

$$\frac{dC_s^{\text{Mie,RL}}}{d\Omega}(\chi_s, m, a_s, \theta) = \chi_s^4 a_s^2 \left| \frac{m^2 - 1}{m^2 + 2} \right|^2 \begin{cases} \frac{1}{2}(1 + \cos^2 \theta) & \text{unpolarised,} \\ 1 & \text{perpendicular } (\perp) \text{ polarised,} \\ \cos^2 \theta & \text{parallel } (\parallel) \text{ polarised.} \end{cases} \quad (14)$$

Since $\chi_s = 2\pi a_s/\lambda_0$, the factor $\chi_s^4 a_s^2$ corresponds to an $a_s^6 \lambda_0^{-4}$ dependence. This expression is used only to describe the small-particle, or Rayleigh-limit, behaviour of particulate Mie scattering. It is useful for checking the small- χ_s limit of the full sphere solution, but not as a substitute for molecular Rayleigh scattering.

A closely related dipole formulation is used for the Rayleigh differential scattering cross section. The magnetic moments of most gas molecules are negligible compared with the electric dipole

moments, so molecular Rayleigh scattering is also purely electric dipole in character (Jackson, 1999). For a dilute gas, the molecular polarisability is related to the relative electric permittivity through the Clausius–Mossotti equation, which at optical wavelengths is commonly written in Lorenz–Lorentz form. With the polarisability defined per molecule, the induced dipole moment is:

$$\mathbf{p} = \varepsilon_0 \frac{3}{N_g} \frac{n_m^2 - 1}{n_m^2 + 2} \mathbf{E}_i, \quad (15)$$

where $N_g = p_g / (k_B T_g)$ is the number of molecules per unit volume. Equation (15) is essentially identical to the small-sphere result in Equation (13), since the number of molecules fitting in a sphere of radius a_s is given by $\frac{4\pi}{3} a_s^3 N_g$. The polarisability is a per-molecule quantity and is independent of density since the refractive index n_m is evaluated at the corresponding density N_g (Sneep & Ubachs, 2005).

For a gas of randomly oriented molecules, the orientationally averaged differential scattering cross section per molecule, including molecular anisotropy through the King correction factor, is given by (Penney, 1969; Jackson, 1999):

$$\frac{dC_g^{\text{Ray}}}{d\Omega}(\lambda_0, n_m, \theta) = \frac{(2\pi/\lambda_0)^4}{16\pi^2} \cdot \left(\frac{3}{N_g} \frac{n_m^2 - 1}{n_m^2 + 2} \right)^2 \cdot \mathcal{F}_k \cdot \begin{cases} \frac{1}{2}(1 + \cos^2 \theta) & \text{unpolarised,} \\ 1 & \perp \text{ -polarised,} \\ \cos^2 \theta & \parallel \text{ -polarised.} \end{cases} \quad (16)$$

In the current work, the gas is assumed to consist of pure nitrogen. The depolarisation factor, or King correction factor, $\mathcal{F}_k$, and refractive index n_m for nitrogen are taken from Naus & Ubachs (2000, eq. 3) and Sneep & Ubachs (2005, eq. 10), respectively. The refractive index is scaled to the local gas number density using the Gladstone–Dale relation with N_g / N_{ref} , where the reference number density is from the ideal gas law at 15 °C and 1 atm.

For particles outside the Rayleigh limit, the arbitrary-size sphere solution is evaluated using the Lorenz–Mie formulation implemented in Matlab by Mätzler (2002a,b), following Bohren & Huffman (1998). The differential scattering cross section for a homogeneous dielectric sphere is given by:

$$\frac{dC_s^{\text{Mie}}}{d\Omega}(\chi_s, m, a_s, \theta) = \left(\frac{a_s}{\chi_s} \right)^2 \begin{cases} \frac{1}{2}(|S_1(\theta)|^2 + |S_2(\theta)|^2) & \text{unpolarised,} \\ |S_1(\theta)|^2 & \perp \text{ -polarised,} \\ |S_2(\theta)|^2 & \parallel \text{ -polarised.} \end{cases} \quad (17)$$

The far-field scattering amplitude functions S_1 and S_2 describe the amplitude and phase of the scattered wave (Bohren & Huffman, 1998; van de Hulst, 1981). For isotropic spherical particles, the angular dependence reduces to these two complex amplitude functions. The dependence on

refractive index m and size parameter χ_s is implicit through the Mie coefficients $a_n(m, \chi_s)$ and $b_n(m, \chi_s)$ used in their series expansion.

The differential cross sections for iron spheres at 532 nm in nitrogen are shown in Figure 6, using $m = 2.8946 + 2.9171i$ from Johnson & Christy (1974), for perpendicular polarised incident light and $\theta = 90^\circ$. The figure compares the full Lorenz–Mie result with the Rayleigh-limit approximation, and also includes the solid-angle-integrated scattering, absorption, and extinction cross sections. These integrated quantities are computed simultaneously within the Mätzler code and complete the theoretical attenuation behaviour of the aerosolised iron spheres considered here. The large-particle extinction cross section approaches twice the geometrical projected area, consistent with the extinction paradox. Following the revised interpretation by Berg et al. (2011), this asymptotic behaviour is understood through Ewald–Oseen cancellation, in which secondary radiation from the particle’s internal field cancels the incident wave inside the particle.

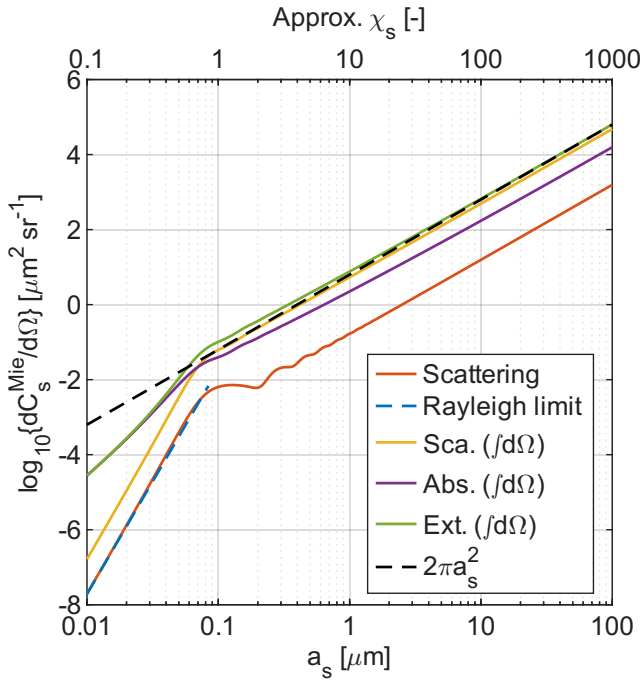


Figure 6. The differential scattering cross section of iron spheres of varying size. Indicated are the particle radius and an approximated size parameter, as χ_s aligns roughly with the order of ten times the particle radius in μm for $\lambda_0 = 532\text{ nm}$. The solid angle integrated scattering, absorption and extinction cross sections are also shown. Note the logarithmic scale on both axes.

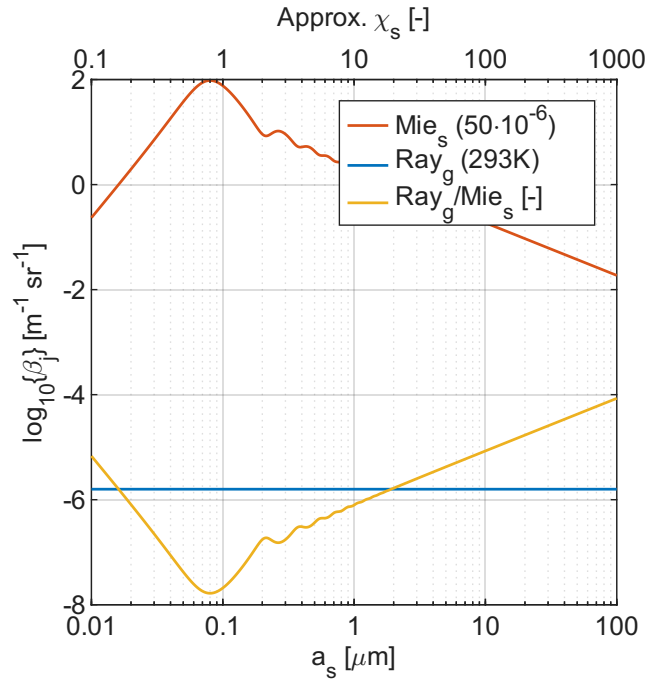


Figure 7. Volume scattering functions for the particulate Mie contribution and the molecular Rayleigh contribution. Iron spheres are evaluated as a function of radius, with the upper axis giving the corresponding approximate size parameter, and at a volume fraction of $50 \cdot 10^{-6}$. Gaseous nitrogen is evaluated at 293 K. The ratio between volume scattering functions is also shown. Note the logarithmic scale on both axes.

2.3.3. Volume scattering functions

With the cross sections defined, the volume scattering functions can be evaluated for both components. For the particulate component, a particle volume fraction f_v is converted to number density through $N_s = f_v/V_s$, with V_s the volume of a single spherical particle. For the gas component, the number density follows from the ideal gas law. Assuming perpendicular polarisation of the incident light, the two volume scattering functions have the dependencies:

$$\beta_s^{\text{Mie}}(\chi_s, m, a_s, \theta, f_v), \quad \beta_g^{\text{Ray}}(\lambda_0, n_m, T_g, p_g). \quad (18)$$

Unless stated otherwise, the remaining calculations assume perpendicular incident polarisation, a 90° scattering angle, nitrogen at atmospheric pressure, iron particles, and an incident wavelength matched to the iodine absorption feature at $\tilde{\nu}_0 = 18787.8 \text{ cm}^{-1}$.

Figure 7 compares the volume scattering functions for a particle volume fraction of $50 \cdot 10^{-6}$ and a gas temperature of 293 K. Additionally, in this graph the ratio of the volume scattering functions $\beta_g^{\text{Ray}} / \beta_s^{\text{Mie}}$ is shown. In this example, Mie scattering always outweighs Rayleigh scattering by several orders of magnitude, a situation that would need to be rectified by the absorption filter in an FRS experiment, as will be discussed in the next section. The same computation can be expanded to a range of particle volume fractions. The resulting number density of particles is shown in Figure 8, where the $50 \cdot 10^{-6}$ line is highlighted by means of a dashed white line. The same logarithmic scaled mapping can be applied to the $\beta_g^{\text{Ray}} / \beta_s^{\text{Mie}}$ ratio, and is shown in Figure 9. This contour provides the order of magnitude by which the particulate contribution must be attenuated for the molecular contribution to become comparable.

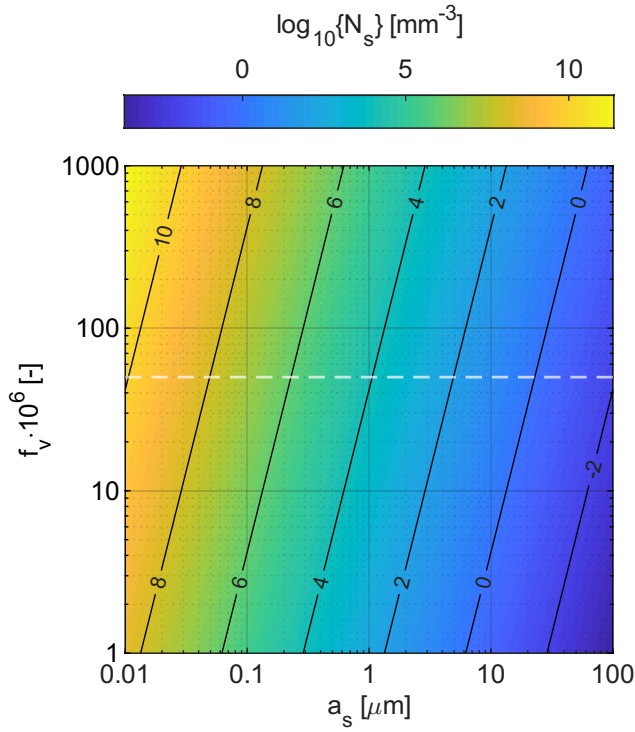


Figure 8. A contour image showing the sphere number density as a function of radius and volume fraction. The dashed white represents the $f_v = 50 \cdot 10^{-6}$ line, utilised in the Mie volume scattering function of Figure 7. Notice the logarithmic scale on both axes, as well as the colour bar and iso-lines.

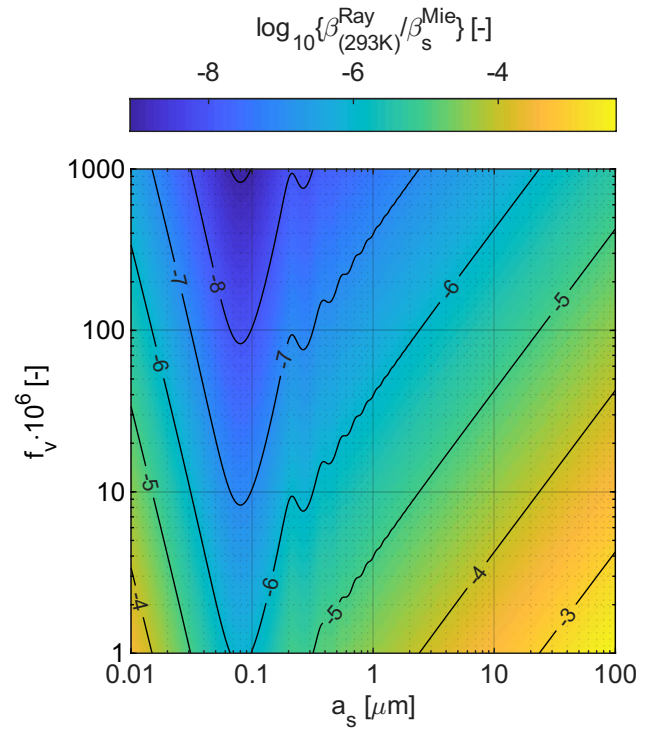


Figure 9. Volume scattering function ratio between the molecular Rayleigh contribution of nitrogen at NTP and the particulate Mie contribution of iron spheres, plotted as a function of sphere radius and aerosol volume fraction. Notice the logarithmic scale on both axes, as well as the colour bar and iso-lines.

2.3.4. Spectral lineshapes and signal integration

The preceding subsections defined the scattering strength per unit volume. The remaining step is to resolve the spectral distribution of each contribution before transmission through the iodine filter. Particulate Mie scattering is assumed to remain spectrally narrow and follows the laser spectrum incident on the probe volume; particle-velocity-induced Doppler shifts are neglected. The molecular Rayleigh spectrum, by contrast, is redistributed over frequency by molecular thermal motion and shaped by density fluctuations that modify the relative strength of the Doppler-shifted frequency components.

In the nomenclature of Young (1982), molecular Rayleigh scattering consists of the Cabannes line and the rotational Raman wings. When the effects of thermal motion and density fluctuations are spectrally resolved, the Cabannes component acquires the Rayleigh–Brillouin structure modelled here. The present model therefore treats the molecular signal as the Rayleigh–Brillouin-resolved Cabannes spectrum of nitrogen, while the rotational Raman wings are not taken into account.

The molecular lineshape is computed with the Pan S6 model (Pan et al., 2002), which is built on the Tenti S6 (Tenti et al., 1974) kinetic model. The incident spectrum downstream of the Fabry–Pérot etalon is convolved with the spontaneous scattering lineshape for nitrogen while conserving the integrated area, so that the differential scattering cross section continues to determine the scattered power. Figure 10 illustrates the resulting redistribution at 293 and 1273 K for nitrogen at atmospheric pressure, using temperature-dependent shear viscosity and thermal conductivity from Sutherland relations in Gu & Ubachs (2014), and the bulk viscosity from Li & Zhang (2025).

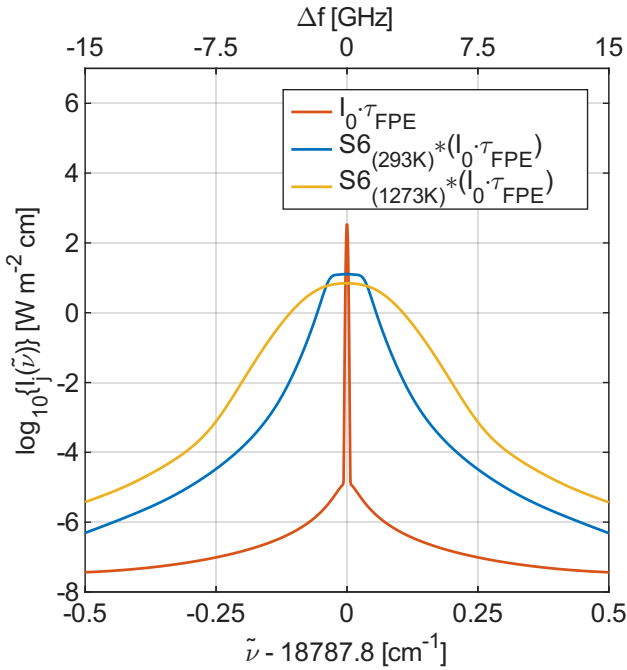


Figure 10. The spectral irradiance of the incident light downstream of the etalon is broadened through a convolution with the Pan S6 molecular lineshape for N_2 at 293 and 1273K.

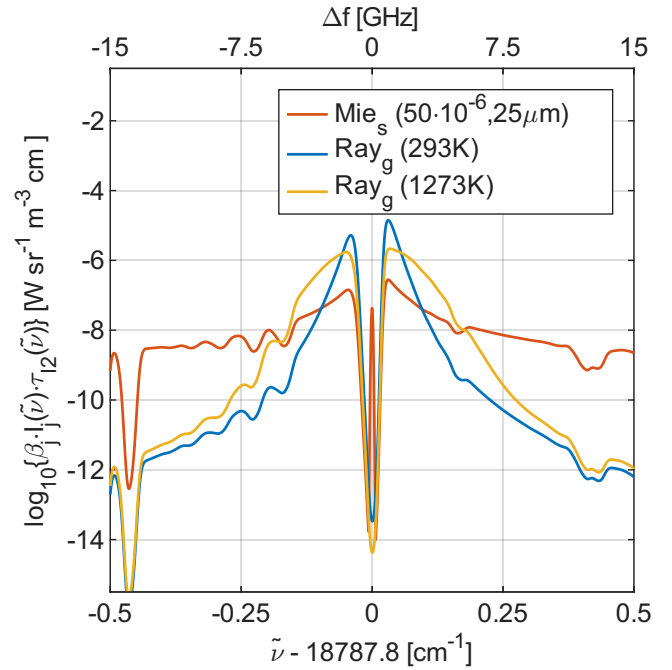


Figure 11. Resolved scattering spectra for the particulate Mie contribution from 25- μm spheres at an aerosol volume fraction of $50 \cdot 10^{-6}$, together with the molecular Rayleigh contribution of N_2 at 293 and 1273K.

These temperature-dependent molecular spectra are directly combined with the corresponding volume scattering functions and transmitted through the iodine transmission profile, together with the spectrally narrow particulate contribution. The resulting filtered spectra are shown in Figure 11 for 25- μm iron particles at 50-ppmv aerosol loading. The iodine absorption line strongly suppresses the narrowband particulate contribution, so that the residual Mie scattering reaching the detector is dominated by the broadband pedestal of the incident laser light. The molecular contribution, in contrast, is distributed over a wider spectral range and is therefore only partially attenuated by the iodine filter.

The power falling upon the detector for each component is obtained from the spectral area of the

corresponding filtered contribution:

$$S_j = \beta_j \int I_j(\tilde{\nu}) \tau_{I_2}(\tilde{\nu}) d\tilde{\nu}. \quad (19)$$

The ratio $S_g^{\text{Ray}} / S_s^{\text{Mie}}$ is used as the feasibility metric for FRS in a particle-laden flow. A value above unity indicates that the filtered molecular signal exceeds the residual particulate signal, and values well above unity provide the required margin for temperature measurements. For the example in Figure 11, which includes the Fabry–Pérot etalon and matches the laser wavelength to the iodine absorption line at 18787.8 cm^{-1} , the ratio gives $S_g^{\text{Ray}} / S_s^{\text{Mie}} = 4.71$ at 293 K. At 1273 K, the ratio decreases to 2.48. This decrease reflects the lower gas number density at higher temperature, but is not governed solely by the conventional T_{ref}/T proportionality in normal unfiltered laser Rayleigh scattering. In FRS, the signal also depends on how the broadened Rayleigh–Brillouin lineshape overlaps with the local iodine absorption lines at and near the chosen wavelength. This effect is shown in Figure 12, where the filtered Rayleigh signal is plotted over a range of temperatures relative to a 293 K reference temperature for a selection of absorption lines.

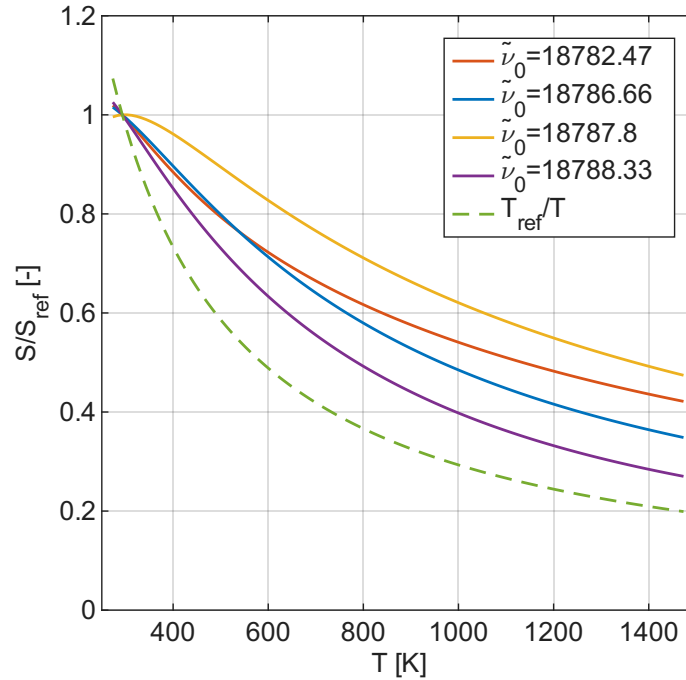


Figure 12. Filtered Rayleigh signal at arbitrary temperature relative to a 293 K-reference temperature, with the laser wavelength matched to different absorption lines. The proportionality T_{ref}/T from the gas number-density scaling is also indicated.

Alternatively, the particle-size and particle-loading dependence is summarised by mapping the complete integrated signal ratio over the same parameter range as in Figures 8 and 9. The results are shown without and with a Fabry–Pérot etalon positioned upstream of the scattering volume in Figures 13a and 13b, respectively.

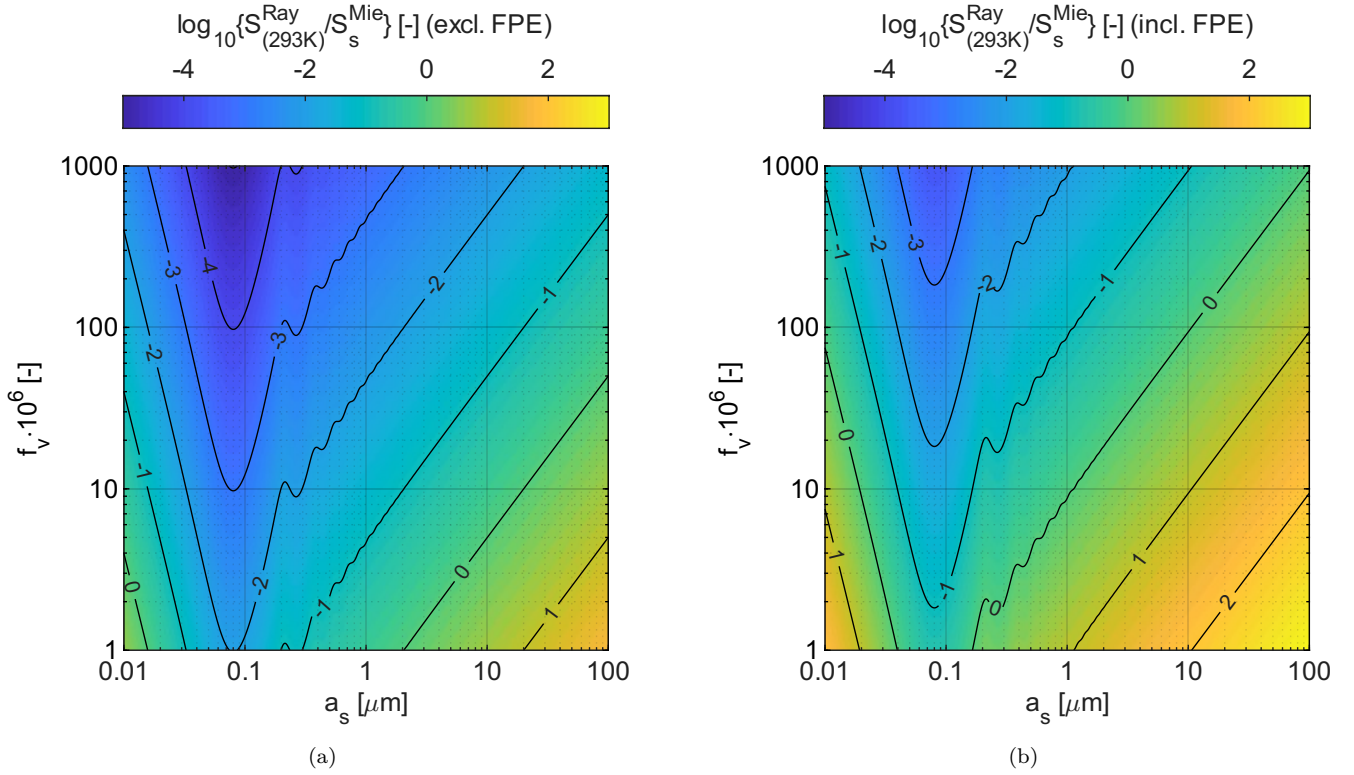


Figure 13. Ratio of the filtered molecular Rayleigh signal and residual particulate Mie signal for a range of particle radius and volume fraction. The colour scale and iso-lines are on a logarithmic scale, so the zero iso-line corresponds to $S_g^{\text{Ray}}/S_s^{\text{Mie}} = 1$. Both axes are logarithmic as well. In a, the spectral purity is that of the laser alone, whereas in b the Fabry–Pérot etalon is added to the optical chain.

At the zero iso-line, $S_g^{\text{Ray}}/S_s^{\text{Mie}} = 1$, so the filtered molecular Rayleigh and residual particulate Mie signals are equal. Adding the Fabry–Pérot etalon reduces the broadband pedestal before the probe volume and increases $S_g^{\text{Ray}}/S_s^{\text{Mie}}$ by 1.27 orders of magnitude with the current settings. The corresponding shift of the zero contour shows that suppressing the broadband pedestal, while maintaining sufficient iodine absorption of the narrowband single-mode component, expands the range of particle conditions for which filtered Rayleigh scattering becomes feasible for temperature measurements within the single-scattering framework used here.

3. Conclusions

A sequential optical-chain model was developed to assess the feasibility of filtered Rayleigh scattering in a particle-laden flow, with example calculations motivated by FRS thermometry in iron-powder flames. The model combines the laser lineshape, optional Fabry–Pérot etalon pre-filtering, iodine absorption, particulate Lorenz–Mie scattering, molecular Rayleigh scattering, and molecular spectral broadening. Reducing the resolved transmitted spectra to the integrated signal ratio $S_g^{\text{Ray}}/S_s^{\text{Mie}}$ provides a practical design metric for identifying operating regions where the filtered molecular signal exceeds the residual particulate signal under single-scattering conditions.

The calculations show that the feasibility boundary depends strongly on particle radius and volume fraction. For the injection-seeded Nd:YAG–I₂ configuration considered here, residual broadband laser-pedestal light can leave a significant Mie contribution after iodine filtering. Adding a Fabry–Pérot etalon upstream of the probe volume reduces this contribution and increases the ratio $S_g^{\text{Ray}}/S_s^{\text{Mie}}$ by 1.27 orders of magnitude for the present settings. The etalon is therefore regarded as a key requirement for particle-laden FRS.

The filtered Rayleigh signal does not follow a simple T_{ref}/T dependence, because the overlap between the Rayleigh–Brillouin lineshape and the iodine absorption spectrum changes with temperature and selected laser wavelength. This makes the choice of laser centre wavelength and absorption feature part of the feasibility challenge. When retaining molecular signal strength is the only objective, a centre wavelength of 18787.8 cm^{-1} is preferred among the absorption lines considered here.

Future work should validate these predicted feasibility regimes experimentally and benchmark the model against published FRS implementations that report seeding conditions. A particularly relevant extension is to include the statistical intermittency of particle contributions in small probe volumes, where discrete-particle fluctuations, rather than the mean particle loading alone, may determine the practical detection limit.

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Nomenclature

a_n, b_n	Lorenz–Mie coefficients [-]
a_s	Particle radius [m]
c	Speed of light [m s^{-1}]
d_{FPE}	Etalon thickness [m]
$dC/d\Omega$	Differential scattering cross section [$\text{m}^2 \text{sr}^{-1}$]
$dP/d\Omega$	Spectral radiant intensity [$\text{W sr}^{-1} (\text{cm}^{-1})^{-1}$]
$\mathbf{E}$	Electric field [V m^{-1}]
f	Frequency [Hz]
f_v	Particle volume fraction [-]
$\mathcal{F}$	Etalon coefficient of finesse [-]
$\mathcal{F}$	Etalon finesse [-]
$\mathcal{F}_k$	King correction factor [-]
$I(\tilde{\nu})$	Spectral irradiance [$\text{W m}^{-2} (\text{cm}^{-1})^{-1}$]
$I_0(\tilde{\nu})$	Laser spectral irradiance [$\text{W m}^{-2} (\text{cm}^{-1})^{-1}$]
k_B	Boltzmann constant [J K^{-1}]
L	Length [m]
m	Relative complex refractive index [-]
N	Number density [m^{-3}]
n_m	Refractive index of surrounding medium [-]
n_{FPE}	Refractive index of etalon medium [-]
n_s	Refractive index of particle material [-]
p	Pressure [Pa]
$\mathbf{p}$	Electric dipole moment [C m]
q_{FPE}	Etalon interference order [-]
$\mathcal{R}$	Etalon reflectivity [-]
S	Integrated signal per unit volume and solid angle [$\text{W sr}^{-1} \text{m}^{-3}$]
S_1, S_2	Lorenz–Mie scattering amplitude functions [-]
SP	Spectral purity [-]
T	Temperature [K]
V	Volume [m^3]
α	Electric polarisability [m^3]
α_w	Lorentzian pedestal weighting factor [-]
β	Volume scattering function [$\text{m}^{-1} \text{sr}^{-1}$]
χ_s	Particle size parameter [-]
δ_{FPE}	Etalon phase difference [rad]
$\Delta\Omega$	Collection solid angle [sr]
ε_0	Electric permittivity of free space [F m^{-1}]

$\Gamma, \tilde{\Gamma}$	Full width at half maximum [Hz or cm^{-1}]
κ_s	Extinction index of particle material [-]
λ	Wavelength [m]
θ	Scattering angle [rad]
θ_{FPE}	Etalon angle of incidence [rad]
$\tilde{\delta}_G$	Gaussian standard deviation [cm^{-1}]
$\tilde{\nu}, \nu$	Wavenumber [cm^{-1} or m^{-1}]
$\tau(\tilde{\nu})$	Spectral transmission [-]

Subscripts and superscripts

0	Quantity evaluated at the laser centre wavelength
I_2	Iodine absorption filter
cell	Vapour filter cell
FPE	Fabry–Pérot etalon
g	Molecular gas component
G	Gaussian
i	Incident
j	Generic scattering component
L	Lorentzian
Mie	Lorenz–Mie particulate scattering
Mie, RL	Rayleigh-limit approximation of Lorenz–Mie scattering
p	Probe
Ray	Molecular Rayleigh scattering
RBS	Rayleigh–Brillouin scattering
ref	Reference condition
s	Particulate or spherical-particle component
stem	Cell side arm
win	Simulation window

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