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Radiative transfer equation accounting for rotational Raman scattering and its solution by the discrete-ordinates method

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ABSTRACT

Rotational Raman scattering of solar light in Earth's atmosphere leads to the filling-in of Fraunhofer and telluric lines observed in the reflected spectrum. The phenomenological derivation of the inelastic radiative transfer equation including rotational Raman scattering is presented. The different forms of the approximate radiative transfer equation with first-order rotational Raman scattering terms are obtained employing the Cabannes, Rayleigh, and Cabannes–Rayleigh scattering models. The solution of these equations is considered in the framework of the discrete-ordinates method using rigorous and approximate approaches to derive particular integrals. An alternative forward-adjoint technique is suggested as well. A detailed description of the model including the exact spectral matching and a binning scheme that significantly speeds up the calculations is given. The considered solution techniques are implemented in the radiative transfer software package SCIATRAN and a specified benchmark setup is presented to enable readers to compare with own results transparently.

1. Introduction

The wavelength dependence of the scattered solar light by Earth's atmosphere is very similar but not identical to the extra-terrestrial solar flux even within spectral ranges free from absorption features of gaseous atmospheric constituents. Indeed, dividing the reflectance spectra by the solar flux, one obtains some spectral structure which highly correlates with Fraunhofer lines demonstrating that the depths of Fraunhofer lines in the solar and reflectance spectra are different. This effect was independently discovered by Shefov in 1959 [1] and Grainger and Ring in 1961 [2]. They have found that the depth of solar Fraunhofer lines in scattered light is a few per cent less than those observed in the direct sunlight.

Subsequent investigations of the so-called Ring- or filling-in effect attributed this phenomenon to a variety of different transspectral processes such as aerosol fluorescence [3], Rayleigh–Brillouin, and rotational Raman scattering [4]. In contrast to elastic scattering processes where the incident photons, after interaction with matter (gas, liquid, and solid), preserve the energy, in transspectral processes the frequency of the photon is shifted to red or blue after interaction. A red shift can be observed when a part of the photon energy is transferred to the interacting matter. The blue shift can be observed when internal energy of the matter is transferred to the photon.

Although most of the photons scattered in Earth's atmosphere are elastically scattered, a fraction of photons can be inelastically scattered by air, predominantly by nitrogen and oxygen molecules. This inelastic scattering process is attributed to Raman scattering. Nowadays scientists concur that rotational Raman scattering (RRS) in Earth's atmosphere explains most of the filling-in of Fraunhofer lines in solar backscatter spectra [5–9].

Rotational Raman scattering can strongly affect the accuracy of trace gas retrievals [7,9,10] from solar backscatter measurements especially in the case of very weak

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gaseous absorbers such as, e.g. iodine monoxide [11] and glyoxal [12]. Furthermore, the filling-in of Fraunhofer lines obtained from the satellite measurements of the backscattered solar light may be used to estimate cloud top pressure [13,14] as well as aerosol properties using both ground-based and satellite observations [15,16]. Therefore, several RT codes have been developed capable of simulating rotational Raman scattering [6,8,9,17,18]. Recently, also the freely available libRadtran radiative transfer package has been extended by a rotational Raman scattering module [19]. Some years ago RRS has been implemented in the radiative transfer model GOMETRAN [9] and later in SCIATRAN [20] for a plane-parallel atmosphere.

The main purpose of this paper is to revise aspects of inelastic radiative transfer theory and to provide a new effective numerical approach which, in the framework of the discrete-ordinates method, allows to solve the radiative transfer equation with first-order rotational Raman scattering terms efficiently. We also expect that this paper could serve as a reference for readers who use SCIATRAN to perform calculations involving RRS.

The outline of the paper is as follows. In Section 2 we present the phenomenological derivation of the radiative transfer equation (RTE) including RRS. Section 3 describes the derivation of the RTE with first-order RRS source terms. The advantages and disadvantages of different approximations are discussed. Three approaches to solve the RTE with first-order RRS source terms in the framework of the discrete-ordinates method are considered in Section 4. In particular, the rigorous and approximate particular integrals method as well as the forward-adjoint approach are discussed. The validity of these methods is considered in Section 5 and a benchmark setup is presented to enable the reader to reproduce the results with SCIATRAN or another radiative transfer model. Speed-up solutions and a discussion of the computational efficiency are presented in Section 6. The paper ends with conclusion and an appendix. The Appendix summarizes analytical expressions for all particular integrals needed to solve the RTE employing rigorous particular integrals method.

2. Phenomenological derivation of the radiative transfer equation

In this section we formulate the radiative transfer equation including rotational Raman scattering. For this purpose we consider the energy balance within an elementary cylinder of the base $d\sigma$ and height ds. Following Vountas et al. [9], the amount of attenuated energy $\mathcal{E}_{att}$ which represents the loss of energy caused by true absorption and scattering in all directions within the cylinder in the wavelength range $d\lambda$ (for unit time period dt) is given as follows:

$$\mathcal{E}_{att} = I(\Omega,\lambda)\sigma_e(\lambda)\, d\Omega\, d\lambda\, d\sigma\, ds, \quad (1)$$

where $I(\Omega,\lambda)$ is the intensity of radiation field at the wavelength λ in the direction characterized by the variable $\Omega := \{\mu,\varphi\}$ which represents a pair of angle variables, $\mu \in [-1,1]$ and $\varphi \in [0,2\pi]$, $\sigma_e(\lambda)$ is the extinction coefficient. Here, μ is the cosine of the polar angle ϑ measured from the positive τ-axis (negative z-axis) and the azimuthal angle, φ, is measured from the positive x-axis towards the positive y-axis. We assume that the right-handed Cartesian coordinate system is used.

The extinction coefficient used in Eq. (1) is written as

$$\sigma_e(\lambda) = \sigma_a(\lambda) + \sigma_p(\lambda) + \beta_{ray}(\lambda), \quad (2)$$

where $\sigma_a(\lambda)$ is the gaseous absorption coefficient, $\sigma_p(\lambda)$ is the extinction coefficient of aerosol and/or cloud particles, $\beta_{ray}(\lambda)$ is the Rayleigh scattering coefficient. As was pointed out by Young [21] one can consider the total Rayleigh scattered energy consisting of two parts. One part is the energy scattered in the spatial domain over all directions without a wavelength shift. Another part of energy is scattered in the spectral domain over a wavelength range which corresponds to the widths of the Stokes- and anti-Stokes bands [22]. Both attenuation processes are already taken into account if the *classical* Rayleigh scattering coefficient $\beta_{ray}(\lambda)$ (see e.g. [21]) is assumed as a component of the extinction coefficient $\sigma_e(\lambda)$.

The wavelength redistribution of energy has to be accounted for properly considering the emitted energy. The energy gain accounting for the rotational Raman scattering can be described by three following processes:

- Rayleigh scattering of energy without shift of the wavelength, i.e., the so-called Cabannes scattering

$$\mathcal{E}_{cab} = \frac{\beta_{cab}(\lambda)}{4\pi}\int_{4\pi} \mathcal{P}_{cab}(\Omega,\Omega')I(\Omega',\lambda)\, d\Omega'\, d\Omega\, d\lambda\, d\sigma\, ds, \quad (3)$$

  where $\beta_{cab}(\lambda)$ and $\mathcal{P}_{cab}(\Omega,\Omega')$ denote the Cabannes scattering coefficient and phase function, respectively (see e.g. [6,7,17,23]);
- contribution of wavelength shifted energy in all Raman lines located in the spectral range $[\lambda, \lambda + d\lambda]$:

$$\mathcal{E}_{ram} = \frac{1}{4\pi}\sum_{j=1}^{L} r(\lambda,\lambda_j) \times \int_{4\pi} \mathcal{P}_{ram}(\Omega,\Omega')I(\Omega',\lambda_j)\, d\Omega'\, d\Omega\, d\lambda\, d\sigma\, ds, \quad (4)$$

  where L is the number of excitation wavelengths, the j-th term in this sum gives the contribution of the energy shifted by the j-th Raman line from the wavelength λ_j into spectral interval $[\lambda, \lambda + d\lambda]$, $r(\lambda,\lambda_j)$ is the RRS coefficient [24,22] (the RRS coefficient and related quantities implemented in the current version of SCIATRAN are summarized in [9]);
- elastic scattering of the energy by aerosol and/or cloud particles:

$$\mathcal{E}_{par} = \frac{\beta_{par}(\lambda)}{4\pi}\int_{4\pi} \mathcal{P}_{par}(\Omega,\Omega')I(\Omega',\lambda)\, d\Omega'\, d\Omega\, d\lambda\, d\sigma\, ds, \quad (5)$$

  where $\beta_{par}(\lambda)$ and $\mathcal{P}_{par}(\Omega,\Omega')$ are the scattering coefficient and phase function of aerosol or cloud particles.

Equating the energy difference between the energy that entered and left the cylinder within the solid angle $d\Omega$ normal to the area of the cylinder in the wavelength range $d\lambda$ given by $d\mathcal{E} = dI(\Omega,\lambda)\, d\Omega\, d\lambda\, d\sigma$ to the difference of the