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# Relation between Raman backscattering from droplets and bulk water: Effect of refractive index dispersion

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ABSTRACT

A theoretical framework is presented that permits investigations of the relation between inelastic backscattering from microparticles and bulk samples of Raman-active materials. It is based on the Lorentz reciprocity theorem and no fundamental restrictions concerning the microparticle shape apply. The approach provides a simple and intuitive explanation for the enhancement of the differential backscattering cross-section in particles in comparison to bulk. The enhancement factor for scattering of water droplets in the diameter range from 0 to 60 µm (vitally important for the *a priori* measurement of liquid water content of warm clouds with spectroscopic Raman lidars) is about a factor of 1.2–1.6 larger (depending on the size of the sphere) than an earlier study has shown. The numerical calculations are extended to 1000 µm and demonstrate that dispersion of the refractive index of water becomes an important factor for spheres larger than 100 µm. The physics of the oscillatory phenomena predicted by the simulations is explained.



## 1. Introduction

The water content of clouds, be it in liquid or frozen form, is one of the key parameters that govern the energy budget of the atmosphere, and thus the weather and by extension the climate of the Earth [1,2]. For this reason accurate measurements of cloud water content are of high importance so that microphysical processes in clouds can be studied and eventually understood better, and numerical weather prediction and climate models may be validated. Over the years, remote sensing has become an integral part of such endeavors for the spatial and temporal coverage it provides. Today, both active and passive instruments are monitoring clouds from space and from the ground continuously, and cloud microphysical products are generated routinely from these observations. However, one should take notice of the fact that these products are often the results of retrieval algorithms based on proxy variables and modeling rather than stemming from direct measurements of the parameter itself, which adds another layer of uncertainty. For instance, in the case of ice water content (IWC), common retrieval techniques employ empirical relations between radar reflectivity (e.g., [3,4]), or lidar extinction coefficient (e.g., [5,6]), and IWC derived from ice particles sampled *in situ* during field campaigns. So, ideally, direct measurement methods should be devised to verify the retrieval techniques. Our objective is to determine liquid water content (LWC) and IWC from lidar measurements *a priori* by utilizing the Raman effect.

The water molecule is Raman-active in all three phases of matter, and Raman scattering by water vapor has been exploited successfully for lidar measurements of atmospheric humidity for a long time (as an early example of an operational water vapor Raman lidar, see [7]). For experimental and methodological reasons, however, Raman lidar studies of the condensed water phases are much more complicated, and despite dedicated efforts over the last years (see the reviews given in [8,9]), *a priori* LWC and IWC measurements have been proven elusive. This is about to change with the advent of spectroscopic water Raman lidars. These instruments allow for the first time direct measurement of the Raman backscatter coefficients of cloud water and ice [9].

Let $\beta$ be the Raman backscatter coefficient of cloud droplets, then

$$\mathrm{LWC} = \frac{K\beta}{\mathrm{d}\sigma_s/\mathrm{d}\Omega}, \qquad (1)$$

where $K$ is a known instrument-specific constant. One can directly obtain LWC from the measurement of $\beta$ provided that $\mathrm{d}\sigma_s/\mathrm{d}\Omega$, the Raman differential backscattering cross-section of a water molecule within a water droplet (subscript 's' stands for sphere) is known. A similar relation applies to IWC, only the numerical

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values of $K$, $\beta$, and $d\sigma/d\Omega$ (being shape dependent) are different. Note, however, that $d\sigma_s/d\Omega$ is not the same as the cross-section $d\sigma_b/d\Omega$ determined in laboratory experiments using bulk samples (subscript 'b' for bulk), but differs from it substantially and exhibits a size dependence as previous studies have shown [10,11].

Let $\eta_s$ be the ratio of the molecular cross-section in a droplet to the one in the bulk water sample, henceforth called the enhancement factor:

$$\eta_s = \frac{d\sigma_s/d\Omega}{d\sigma_b/d\Omega}, \quad (2)$$

then Eq. (1) can be rewritten as:

$$\mathrm{LWC} = \frac{K\beta}{\eta_s \, d\sigma_b/d\Omega}. \quad (3)$$

So in order to obtain LWC *a priori*, we have to determine the Raman differential backscattering cross-section of a water molecule in a macrosample and the magnitude of the size-dependent enhancement factor. In a previous publication, we have obtained $d\sigma_b/d\Omega$ with high accuracy [12], the subject of the present paper is the investigation of $\eta_s$. Because the situation is even more complicated for ice due to the enhancement factor being dependent on the shape of the ice particle [13,14], we focus here mostly on the liquid phase. The enhancement factor for ice particles will be discussed in a follow-up article.

Incidentally, we point out that a study of the enhancement factor of water droplets was published previously [10] which, however, was restricted to relatively small size parameters and left some questions unaddressed. Thus our motivation has been threefold: (1) Find a simple and intuitive explanation for the enhancement of the molecular Raman backscattering cross-section in water droplets in comparison to bulk samples. (2) Determine the magnitude of $\eta_s$. Because any error in $\eta_s$ directly affects LWC results, this knowledge is crucial. (3) Extend the droplet size range to diameters of drizzle and small rain drops for which a spherical shape may still be assumed, and explore the dependence of $\eta_s$ on size.

The article is organized as follows. In Section 2, the theory of our model is described in detail. We have followed a new approach and have applied the Lorentz reciprocity theorem to the analysis of Raman scattering by particles. The numerical results are presented and discussed in Section 3. Conclusions are drawn and an outlook is given in Section 4.

## 2. Theory

The following theory is basic and is not limited to the case of spherical liquid droplets. To evaluate the value of $\eta$, we use a new approach based on Lorentz reciprocity theorem [15] which states that for any volume and its enclosing surface $S$ the following relation between the volume and surface integrals

$$\int [\vec{J}_1\vec{E}_2 - \vec{J}_2\vec{E}_1]\mathrm{d}V = \oint_S [\vec{E}_1 \times \vec{H}_2 - \vec{E}_2 \times \vec{H}_1]\mathrm{d}\vec{S} \quad (4)$$

holds for two sinusoidal current densities $\vec{J}_1$ and $\vec{J}_2$ oscillating at the same frequency and generating the electromagnetic fields $\vec{E}_1, \vec{H}_1$ and $\vec{E}_2, \vec{H}_2$. For a particular case of $\vec{J}_1$ and $\vec{J}_2$ being the currents of two point dipoles and the volume covering the whole space, the surface integral vanishes and the theorem simplifies to

$$\vec{\mu}\vec{E}^{(d)} = \vec{d}\vec{E}^{(\mu)} \quad (5)$$

where $\vec{E}^{(d)}$ is the field created by a point dipole $\vec{d}$ at the location of point dipole $\vec{\mu}$ and $\vec{E}^{(\mu)}$ is the field created by $\vec{\mu}$ at the location of $\vec{d}$.

Suppose that the point electrical dipole $\vec{\mu}$ is immersed in a dielectric of an arbitrary shape. The dielectric material occupies volume $V$. Both dipoles oscillate at angular frequency $\omega'$. We assume a large distance between the two dipoles (much larger than the size of $V$ and the wavelength of the wave). Without a loss of generality, we can also assume that $\vec{d}'$ is oriented along $x$-axis of the coordinate system and consider a wave radiated by this dipole propagating in $z$-direction towards $\vec{\mu}$. At a large distance from $\vec{d}'$, the electromagnetic wave emitted by $\vec{d}'$ can be treated as a plane $x$-polarized wave (this wave is considered plane within $V$). The electrical field of this (*pumping*) wave reads $E_0 \exp(k'z - i\omega' t)$, where $E_0 \propto d'$.

When the *pumping* wave interacts with the dielectric volume, the internal field (inside the volume) can be presented as a vector field $\vec{E}_i^{(x)}(x, y, z, \omega')$, where we drop the time-dependent factor $\exp(-i\omega' t)$ and the superscript indicates that the internal field is calculated for the case of a plain, $x$-polarized incident wave. Suppose that $(x, y, z)$ is the location of the dipole $\vec{\mu}$ which is induced by $\vec{E}_i^{(x)}$. In the simplest case of Raman scattering, $\vec{\mu} = \alpha\vec{E}_i^{(x)}(x, y, z, \omega')$ with $\alpha$ being polarizability but it oscillates with angular frequency $\omega$. The field produced by this dipole is the *scattered* wave and can be obtained from Eq. (5) by considering an auxiliary dipole $\vec{d}$. Generally, the angular coordinates of this dipole can be arbitrary, but here we take a practically important case of backscattering when the location of $\vec{d}$ coincides with $\vec{d}'$. For simplicity it is assumed that $|\vec{d}| = |\vec{d}'|$. Vector $\vec{d}$ can be either parallel or perpendicular to $\vec{d}'$. In the case of $\vec{d} \parallel \vec{d}'$, one gets $\alpha\vec{E}_i^{(x)}(x, y, z, \omega)\vec{E}_i^{(x)}(x, y, z, \omega') = dE_x^{(\mu)}$. The projection of the scattered field on $y$-axis can be obtained by considering $\vec{d} \perp \vec{d}'$ which results in $\alpha\vec{E}_i^{(y)}(x, y, z, \omega)\vec{E}_i^{(x)}(x, y, z, \omega') = dE_y^{(\mu)}$.

If there are many *incoherent* induced dipoles homogeneously distributed over the entire volume $V$, then one can get the total power radiated by these dipoles in the direction to the dipole $\vec{d}$ by integration. The differential $x$-polarized backscattering cross-section per dipole is the radiant intensity of the scattered wave (proportional to $|E_x^{(\mu)}|^2$) divided by the intensity (irradiance) of the pumping wave (proportional to $|E_0|^2$) and similar for the $y$-polarized scattering. Thus, one gets

$$\frac{d\sigma_V^{(x)}}{d\Omega} = \frac{Y|\alpha|^2}{V|\vec{E}_0|^4} \int_V \left|\vec{E}_i^{(x)}(x, y, z, \omega)\vec{E}_i^{(x)}\left(x, y, z, \omega'\right)\right|^2 dV \quad (6)$$

and

$$\frac{d\sigma_V^{(y)}}{d\Omega} = \frac{Y|\alpha|^2}{V|\vec{E}_0|^4} \int_V \left|\vec{E}_i^{(y)}(x, y, z, \omega)\vec{E}_i^{(x)}\left(x, y, z, \omega'\right)\right|^2 dV \quad (7)$$

where $Y$ absorbs all the constant factors such as speed of light in vacuum, concentration of dipoles etc. This constant also includes a factor dependent on the units, photon/(s sr) or W/sr used for the radiant intensity. The value of the total backscattering cross-section (a common case of lidar measurements is integration of scattering over both polarizations) can be obtained as a sum of the two values:

$$\frac{d\sigma_V}{d\Omega} = \frac{d\sigma_V^{(x)}}{d\Omega} + \frac{d\sigma_V^{(y)}}{d\Omega}. \quad (8)$$

### 2.1. Bulk Raman scattering

First, we apply Eqs. (6)–(8) to the case of bulk scattering. In such a case the dielectric is a large volume (theoretically a half-space) and has a plain interface with air but the scattering is collected from a volume small in comparison to the size of the bulk sample (see Fig. 1). In practice, this volume is defined by the details of the experimental setup. The internal field inside the bulk $E_i^{(x)}(x, y, z, \omega)$ is uniform and in accordance with Fresnel's formula reads

$$E_i^{(x)}(x, y, z, \omega) = \frac{2}{n+1} E_0 \exp(ikz), \quad (9)$$