A SPONTANEOUS RAYLEIGH-BRILLOUIN SCATTERING EXPERIMENT FOR THE CHARACTERIZATION OF ATMOSPHERIC LIDAR BACKSCATTER

Executive Summary

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Motivation for the study and introduction

The present study is intimately linked to future missions of the European Space Agency. The immediate connection is to the ADM-Aeolus mission aiming to produce a velocity (wind) profile of vertical layers in the Earth atmosphere *on a global scale*. This is pursued by *active* remote sensing, i.e. by measuring the spectral profile of the back-scattered light from an ultraviolet laser on board of the satellite.

In the recent past it was noted that the fact that expected molecular scattering functions are not just Gaussian profiles and may influence the Doppler measurements and impact the wind profile analysis. In particular acoustic phenomena known to produce the characteristic Brillouin side-wings on the Doppler profile have a strong effect. This was identified as a major problem in a previous study (the ILIAD report[1]) and it was estimated that neglecting Brillouin effect might result in errors in the radial wind measurement of up to 10% in several cases. These estimates were made on the basis of models known in the literature since the 1970s as the TENTI models. These TENTI models had only been tested for a few measurement configurations and for a very small subspace of all possible gases, pressures and mixtures. It had not been tested for air under the various atmospheric conditions. Hence the goal of the present project was defined: measuring the spontaneous Rayleigh-Brillouin (RB) scattering profile and comparing them to both versions of the TENTI-models (the so-called TENTI S6 and TENTI S7 varieties) in conditions relevant for upcoming ESA LIDAR missions. The objectives of the present study, as defined in the contract, were:

- Quantify the contribution of RB scattering to LIDAR molecular backscatter in a well-defined laboratory experiment.
- Validate the performance of the test equipment by reproducing the measurements of spontaneous RB scattering in N_2 as given by literature.
- Validate the TENTI (S6 and S7) model for atmospheric gas mixtures representing the Earth's atmosphere and assess the necessity of applying refinements to it.
- Make the necessary improvements of the TENTI (S6 and S7) model.
- Make recommendations for the use of the model in the Earth Explorer Core Mission and post-EPS Doppler Wind LIDAR retrieval algorithms.

We have performed measurements of the molecular scattering profile by investigating *coherent* Rayleigh-Brillouin spectroscopy in addition to the *spontaneous* RB measurements. Specifically for the central goal of understanding the TENTI-models and testing them in an as broad as possible parameter space, a second and independent technique is very valuable. While there had been a strong activity on spontaneous RB scattering of gases in the 1960s and 1970s, also leading to the formulation of the TENTI-formalism, the subject was left aside since then. The development of coherent RB scattering took place in the last decade. It was shown that the scattering profiles were essentially different (between spontaneous and coherent RB), but they could be related in a similar manner to the TENTI models.

[1] *ILIAD - Impact of Line Shape on Wind Measurements and Correction methods*
C. Loth, P. H. Flamant, A. Dabas, M.-L. Denneulin, A. Dolfi-Bouteyre, A. Garnier and D. Rees, *ESA Contract No 18334*, (2005) 124p.

Overview of the results

We have measured the line shapes of the SRBS returns from different gases (N_2, O_2) and gas mixtures (air) at various pressures (see Tables).

Table 1: Measurement and models (Tenti S6 and S7), for measurements performed at different pressures and ambient temperature.

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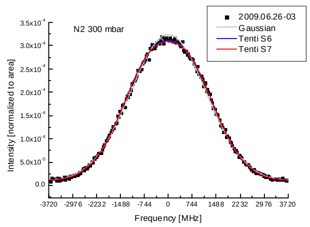
<td>
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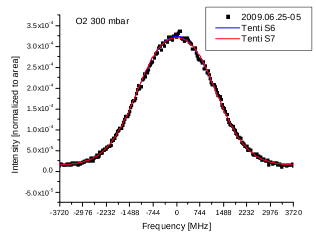
<td>
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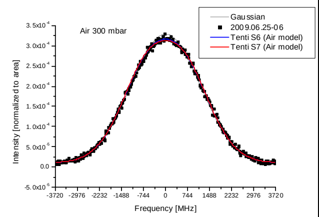
<td>
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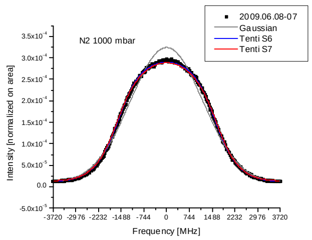
<td>
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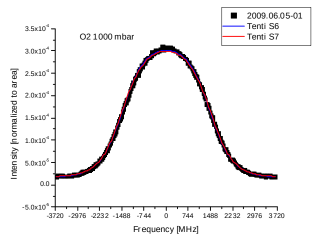
<td>
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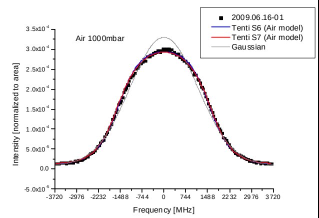
<td>
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Table 2: Deviations model-measurement in percentage of the maximum signal of the model, for measurements performed at different pressures and ambient temperature.

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<tr><th>P [bar]</th><th>N2</th><th>O2</th><th>Air</th></tr>
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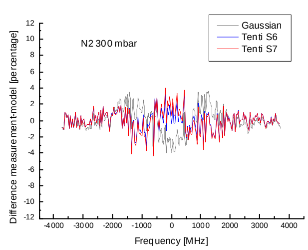
<td>
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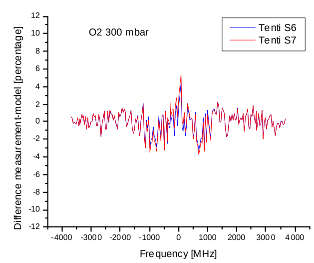
<td>
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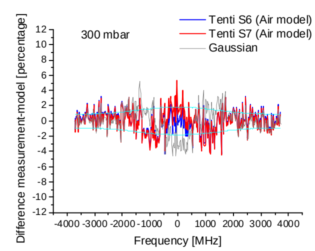
<td>
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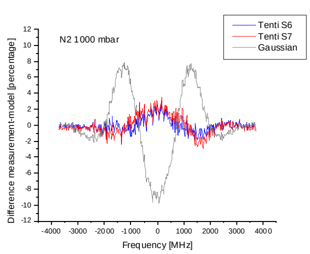
<td>
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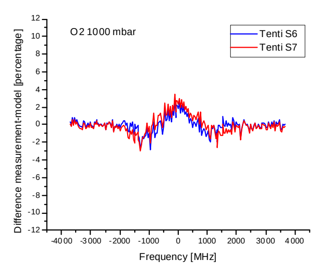
<td>
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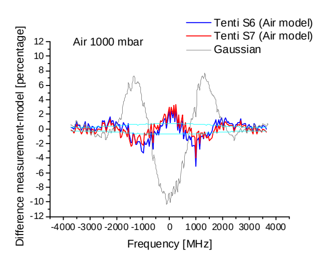
<td>
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Humidity measurements were performed for air at 1040 mbar (sea level pressure). There were three measurements: i) at 0% humidity, ii) when the humidity was increasing, iii) and when there was 100% humidity (saturated water vapor) in the cell.

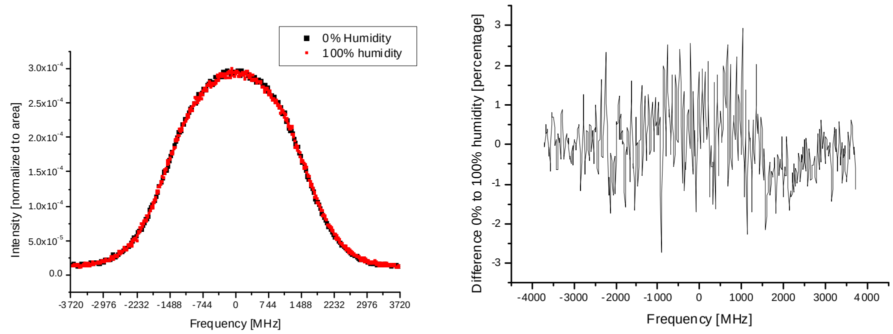

Figure 1: Measurements of 0% and 100% humidity alone for better comparison. The plot on the right is the difference between 0% humidity and 100% humidity, divided by the maximum value of the measurement with 0% humidity. It can be concluded that the humidity effect in these conditions is not detectable. There are no striking differences between measurements with or without water vapor. The residual plot shows a scattering of points around zero.

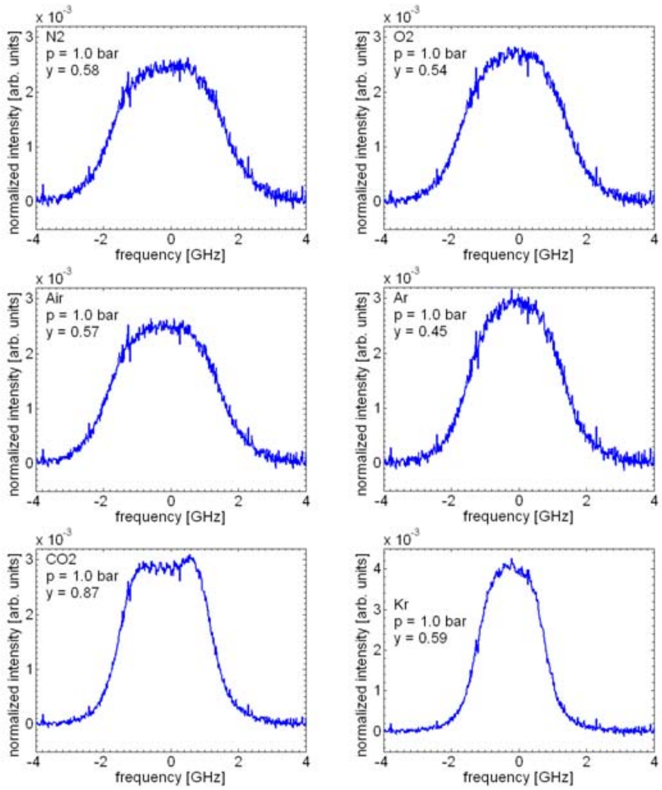

Figure 2: Experimental results on Coherent Rayleigh Brillouin scattering at 532 nm for various pressures in N_2, O_2, Air, Ar, CO_2 and Kr.

CRBS spectra have been measured in N_2, O_2, air, CO_2, Ar, and Kr at pressures of 1, 2 and 3 bar. Above a small sample of the experimental results is shown.

Conclusions and recommendations

The combination of the SRBS and CRBS experiments allows for an independent test of the Tenti-models describing RB-scattering and they provide the possibility to probe the parameter space of the Tenti models in a wide range. Both experiments were successful in that they produced the **highest quality (highest signal-to-noise) RB-profiles of gaseous species measured so far**, both for the spontaneous as for the coherent case.

Measurements were performed in the pressure ranges between 300 mbar and 3 bar for the spontaneous RB setup and between 1 and 3 bar for the coherent RB setup. Both setups measured scattering in dry air, pure N_2 and pure O_2 gases, as well as Kr. It was decided to extend the pressure range above the atmospherically relevant value of 1 bar, because these higher pressures are closer to the hydrodynamic regime, where the Brillouin side peaks are more pronounced. Hence, in this part of the parameter space the deviations on the profile from a pure Gaussian can be better modeled, and the Tenti model better tested.

The possible effect of water vapor was addressed in a spontaneous RB-measurement, while making a comparison between line shapes for dry air and humid air (i.e. air fully saturated with water vapor) at 1 bar. This resulted in a non-observable difference, which may be quantified as an effect by the **water vapor content contributing less than 0.5% to the scattering intensity**.

A **new FORTRAN code** was written for both the Tenti-6 and Tenti-7 models, which is now applicable for **all gases, for all scattering geometries and for both the spontaneous and the coherent RB** scattering cases. This code contains the transport coefficients (best known so far) of several gases, while for air also the possibility of temperature dependence of the transport coefficients is implemented. The Tenti model is from its principles only adaptable for single species gases, but it was made applicable to air, by treating air as consisting of a single-species molecule with some macroscopic transport coefficients as know from literature. The code is easily implemented to produce model profiles for any RB-condition, where it can be compared with a measured profile. The program is user-friendly, while allowing for the treatment all gases (just the database of constants must be filled), and maintaining a high processing speed that would allow for its use online with the satellite measurements.

Comparisons between the high quality experimental data with the Tenti-codes led to the following conclusions:

- The Tenti-6 model describes the observations for air, N_2 and O_2, over a wide parameter space to within 2%, at least for the atmospherically relevant conditions of pressure.
- Two refinements have been performed in the treatment of the model: to allow the use of the air transport coefficients (treating air as a “single species”) and the “fit” of the bulk viscosity value. Overall, the model (Tenti S6) matches the air measurements better with a bulk viscosity value that it is roughly double of the literature value.

Extra refinement of the Tenti model is at this stage not a workable option. The model is the current “best description” of the RB-scattering process, and it has a solid basis in physics. Numerical adaptation of the Tenti model is not an option either (perhaps it can for a determination of the bulk viscosity parameter as discussed above). We stress also that more precise experimental data on some of the transport coefficients would be needed, in particular as a function of temperature. A direct measurement of the bulk viscosity at optical frequencies would also be very useful to unravel the riddle of the RB-scattering profile.

For the moment it must be concluded that the **Tenti-6 model yields the best description** and is at the same time rooted in physical insight in the processes underlying the scattering profile. Deviations at the 2% level are found when subjecting this model to experimental tests under various conditions, and these deviations are beyond the possibility of modeling. So the current situation is that there exists an optimum line shape (Tenti-6) and there is quantitative knowledge on the deviation of the experimental data from this line shape.

As is shown in this study, small uncertainties in the knowledge of the spectral shape of the Rayleigh-Brillouin backscattered are of importance to ESA's Lidar missions. For the ADM-Aeolus Lidar mission in particular, such uncertainties may lead to errors in the Aeolus-retrieved-line-of-sight-winds of a few m/s.

The residuals between the modelled (Tenti) and "measured" (Tenti + measurement residuals) line shapes that were projected to the Aeolus configuration, show that the uncertainties in the Tenti S6 RB model are generally much smaller than errors made when assuming a purely Gaussian molecular motion PDF. When assuming that the "measured" (Tenti + residual) line shape is correct, most simulations show a bias of around 1 m/s and a response slope error of 0.2-0.9% in the Aeolus retrievals than when using the Tenti S6 RBS line shapes.

The temperature and pressure dependency of the line-of-sight wind errors have been studied before in the ILIAD study. The results from this study are consistent with the current results since the same line-of-sight wind deviations, up to about 8 m/s were found at surface pressures and high input winds, when the wind retrieval was done using the Gaussian spectral shape.

Since the Level-2B processing stage uses a look-up-table to invert the response of the FP spectrometer to wind, the table can be simply exchanged for a new one without much effort, as soon as a new and better model of the molecular motion spectrum would become available. Therefore efforts to improve the model can be continued until and even after the launch of ADM-Aeolus (validation from space).

The present study has delivered a wide variety of scattering profiles from both RB-spectrometers that were specifically developed for this ESA study. The 18-month duration was too short to cover the entire parameter space. Some important issues dealing with the understanding of the RB-profiles are therefore still open, and could be measured without major difficulty now that the time-consuming process of constructing the spectrometers has been overcome. Interesting open issues are: *Wavelength* effects, *Angle* effects, *Temperature* effects, *Polarization* effects.

In addition we mention two further possibilities to use the novel constructed spectrometers and to extend the knowledge of RB scattering profiles in a scientifically important domain. Scattering profiles of carbon dioxide are of major relevance for the atmosphere of Venus, which is subject to existing studies, and possibly also of future ESA missions. Also, the Tenti model has not been tested for molecules with specific attractive forces, as for example occurring in polar gases. This could be tested by probing scattering profiles in high density gases such as CO, ammonia, or NO.