Spontaneous Rayleigh-Brillouin Scattering in Molecular Gases

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Summary

In this thesis it is shown that high signal-to-noise ratio Rayleigh-Brillouin (RB) scattering profiles can be measured at high spectral resolution. The RB-spectra, obtained under a variety of measurement parameters, such as pressure, temperature, incident wavelength and scattering angle, form a powerful tool for studying the thermal dynamic and gas transport properties associated with the collective motion and relaxation phenomena in the gas. Different models have been proposed to describe this complex motion involving acoustic excitation and relaxation. Four different models, application in certain conditions, are experimentally tested and relevant gas transport parameters are derived. In particular the bulk viscosity, one of the elusive parameter is determined.

In **chapter 2**, two measurement setups are described: a complex one using UV-light and simpler one using green light. The optical components and their characteristics are described in detail and the method of data acquisition and processing are explained.

In **chapter 3**, the bulk viscosity of SF_6 is obtained through comparing the measured RB-scattering data under varying conditions with three models: the Tenti-S6 model, the rough-sphere model and Hammond-Wiggins hydrodynamic model. For the scattering approach Knudsen regime, the hydrodynamic model is not suitable, and we suppose the residuals between these three models and experimental data are from the using of the ideal gas law for the models.

In **chapter 4**, RB-scattering spectra of N_2O are compared with the Tenti-S6 model, the Grad's six-moment model, the Hammond-Wiggins hydrodynamic model and the rough-sphere model. From the results, the bulk viscosity is found to be pressure dependent. The Grad's six-moment model has a similarly good performance as the Tenti-S6 model. The rough-sphere model is demonstrated to not be applicable in view of the non-spherical and non-symmetrical geometric structure of the N_2O gas.

Chapter 5 shows another study of RB-scattering of a linear structure of the symmetric molecule, CO_2. The spectra of CO_2 are measured at different pressures and temperatures. The bulk viscosity of CO_2 obtained

from an RB-scattering experiment is found to be four orders of magnitude smaller than that obtained using the method of sound absorption. The comparison of scattered light spectra to kinetic and hydrodynamic models shows that this dramatic frequency dependence of the bulk viscosity is due to the (gradual) cessation of vibrational relaxation. There is no significant temperature dependence.

In **chapter 6**, the RB-scattering of binary mixture gases is investigated. A large amount of data has been collected for mixtures of gases, where one component is a strong scatterer, while a second component acts as 'spectator', only influencing the collisional dynamics. In case of SF_6-He and CO_2-He binary mixtures, the RB-scattering intensity is entirely produced by the strongly polarizable SF_6 and CO_2. Nevertheless, the spectral profiles are strongly influenced by the additional of the atoms, that influence the motion and collisional dynamic of the SF_6 and CO_2 molecules. In addition, SF_6-D_2, CO_2-D_2, SF_6-H_2, SF_6-CH_4 as well as CO_2-CH_4 mixtures are also studied. A full quantitative treatment and modeling the data is presently under way.

The primary perspective of the present investigations was to measure high quality Rayleiigh-Brillouin scattering spectra with the twofold aim to (i) produce scattering profiles to compare with light scattering media under realistic conditions, and (ii) to determine thermodynamic gas transport coefficients. As for the applicability of the data the investigation of RB scattering in CO_2 is to be mentioned. Carbon dioxide is a prominent greenhouse gas and its capture, transport, storage, and conversion is of relevance for a sustainable environment. Accurate knowledge of the thermal dynamic properties of CO_2 gas, and of gas mixtures containing CO_2, is therefore of relevance. In addition, measurement of its light scattering properties can later support the detection and quantification of CO_2-gas during transport and processing. Light scattering might be applied as an alternative for absorption or emission spectroscopy for detecting species. Also the measured profiles might be of relevance for lidar detection of planetary atmospheres where CO_2 is a major component, like on Venus and Mars. Possibly such light scattering results may be of future relevance for the study of exoplanets. For the measurements on other gases or mixtures thereof the relevance leans more toward finding proper descriptions of thermodynamic and statistical mechanics properties of complex ensembles of particles. The study of gas mixtures has been explored in the past but it is for the first time that systematic studies yielding such accurate data are performed. The acquired experimental data form a starting point for extended modeling, perhaps opening up a new direction in statistical mechanics.