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1 Introduction

When an electromagnetic wave encounters a small obstacle, it is partially deflected from its original direction. This process is called scattering. The effects that occur depend on the ratio between the wavelength and the size of the obstacle. Although Gustav Mie provided a universal description in 1908, the special case where the obstacle size is small compared to the wavelength was already described by Lord Rayleigh in 1871. This case is still referred to as Rayleigh scattering. A characteristic of Rayleigh scattering is the strong dependence on the wavelength: the fraction of scattered light is proportional to the fourth power of the frequency of the light.

A prominent example of Rayleigh scattering is the scattering of sunlight by molecules in the Earth's atmosphere. Although the blue portion of sunlight is relatively small, the wavelength dependence of Rayleigh scattering is so strong that the scattered light is dominated by the blue portion. This is why the sky appears blue when one does not look directly at the Sun. Conversely, the intensity of unscattered light is reduced most strongly in the high-frequency (blue) portion, and least in the red portion. During the day, this second effect is hardly noticeable because the Earth's atmosphere is relatively thin and, overall (for nearly perpendicular light incidence), only a small fraction of the sunlight is scattered. However, where the light passes through the atmosphere tangentially, the path length through the atmosphere is much longer, and the fraction of scattered light is therefore much greater. This is why the Sun appears not only dimmer (allowing direct observation with the naked eye) but also redder shortly after sunrise and just before sunset. In this case, the red portion of unscattered light is less attenuated than the other spectral portions.

2 Theory

2.1 Rayleigh Scattering

If the scattering object is a single atom or molecule that is small compared to the wavelength of light, it can be approximated as an oriented dipole. The electrons oscillate in the direction and at the frequency of the electric field, i.e., perpendicular to the direction of wave propagation. This model describes the scattered light as the radiation emitted by this dipole. The angular dependence of the intensity of this radiation is given by

$$I(\theta) \propto \sin^2(\theta),$$

while the total scattered intensity I_s (integrated over all solid angles) is proportional to the intensity I_i of the incoming wave:

$$P_s = \sigma(\lambda) \cdot I_i = \sigma(\lambda),$$

where the wavelength-dependent cross-section is

$$\sigma(\lambda) \propto \frac{1}{\lambda^4}.$$

In experiments, it is easier, because we study scattering not on a single molecule but on a gas volume $V = L \cdot A$ (with L: length in the propagation direction, A: cross-sectional area). Given a particle density N, the total number of particles in the volume is

$$n = N \cdot V = N \cdot A \cdot L.$$

A homogeneous light beam of cross-sectional area A will undergo n scattering events as it passes through this volume. With each scattering event, the power of the unscattered light decreases to

$$P' = P - P_s = P - \sigma \cdot I = P - \frac{\sigma \cdot P}{A} = (1 - \frac{\sigma}{A}) \cdot P,$$

where P is the power before scattering. After N successive scattering events, the power decreases to

$$P' = (1 - \frac{\sigma}{A})^n \cdot P \approx (1 - \frac{n \cdot \sigma}{A}) \cdot P = (1 - N \cdot L \cdot \sigma) \cdot P = (1 - \beta \cdot L) \cdot P,$$

with $\beta := N \cdot \sigma$. The corresponding intensity is therefore

$$I' = (1 - \beta \cdot L) \cdot I. \tag{1}$$

A more detailed consideration of dipole radiation yields

$$\beta = \frac{8\pi^3 (n^2 - 1)^2}{3N\lambda^4}, \tag{2}$$

where n is the refractive index. Since n depends on the concentration, this resolves the apparent contradiction that N appears in the denominator of Equation (2), even though β increases with N.

2.2 Cavity Ring-Down Spectroscopy

As mentioned in the introduction, the intensity reduction caused by Rayleigh scattering in air is so small that a length of several kilometers is required to observe a significant effect. Experimentally, this can be achieved with a cavity, i.e., a space where light passes multiple times with the help of two mirrors. The light must first be directed perpendicularly into the cavity and, after many passes, extracted for intensity measurement. For this purpose, partially transmissive mirrors with high reflectivity are used. Instead of a continuous light beam, a short light pulse is used, allowing the number of light passes through the cavity to be determined using time. For k passes of length L, the light requires the time $t = k \cdot L/c$ (with c: speed of light), so

$$k = \frac{c \cdot t}{L}. \tag{3}$$

At each reflection on the partially transmissive mirror, a constant fraction $T = 1 - R$ of the light leaves the cavity. The intensity $I_T(t) = T \cdot I(t)$ leaving the cavity at time t is therefore proportional to the intensity remaining in the cavity at that time. At the same time, the remaining intensity decreases by the transmitted intensity after each pass, i.e., the remaining intensity is reduced to $I' = R \cdot I$ after each pass. This holds regardless of whether the cavity is empty or filled with gas. If it is gas-filled, the intensity has already been reduced by scattering according to Equation (1), so the intensity remaining after each pass is

$$I' = R \cdot (1 - \beta \cdot L) \cdot I,$$

where I is the intensity at the start of the pass. For k passes, the intensity is therefore

$$I(k) = (R \cdot (1 - \beta \cdot L))^k \cdot I_0,$$

or using Equation (3),

$$I(t) = (R \cdot (1 - -\beta \cdot L))^{c \cdot t/L} \cdot I_0 = \exp\left(\frac{c \cdot t}{L} \cdot \ln(R \cdot (1 - \beta \cdot L))\right).$$

This simplifies to

$$I(t) = \exp\left(-\frac{t}{\tau}\right),$$

with

$$\tau = -\frac{L}{c \cdot \ln(R \cdot (1 - \beta \cdot L))} = -\frac{L}{c \cdot (\ln(R) + \ln(1 - \beta \cdot L))}.$$

$$\tau \approx \frac{L}{c \cdot (1 - R + \beta \cdot L)}.$$

By performing measurements once with an evacuated cavity ($\beta = 0$) and once with a gas-filled cavity, the unknown quantity R can be eliminated, and β can be determined:

$$\beta = \frac{1}{c}\left(\frac{1}{\tau} - \frac{1}{\tau_0}\right), \tag{4}$$

where τ is the decay constant for the gas-filled cavity and τ_0 is the decay constant for the evacuated cavity.

3 Description of the Experimental Setup and Technique

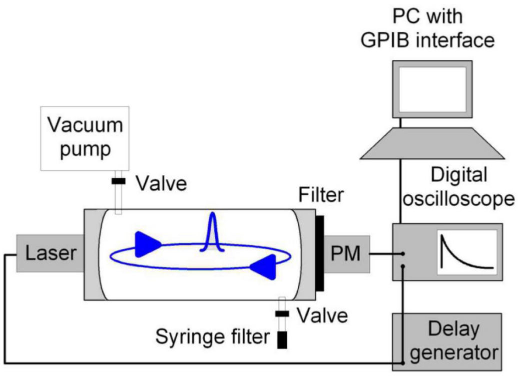

Figure 1: experimental set up

The experimental setup consists of the following components:

1. **Laser Source**: A pulsed laser with $\lambda = 405\,\text{nm}$ is externally triggered by a delay generator.
2. **Optical Cavity**: Two highly reflective spherical mirrors ($R > 99.98\%$) form a resonator cavity with a separation of 50 cm. The cavity can be evacuated to measure the CRD time τ_0 without scattering.
3. **Detector**: A photomultiplier tube (PMT) detects the transmitted light intensity after it passes through a bandpass filter to reduce background noise.
4. **Data Acquisition**: The signal from the PMT is recorded on a digital oscilloscope and transmitted to a computer for further analysis.
5. **Auxiliary Components**: A vacuum pump and a barometer are used to create and monitor the evacuated conditions, while a syringe filter prevents large particles from entering the cavity.

The procedure begins with aligning the laser beam parallel to the cavity using a visible green laser ($\lambda = 532\,\text{nm}$) for calibration. After alignment, the pulsed laser is used to measure the intensity decay $I(t)$ alternately with the cavity filled with air and under vacuum. These measurements allow the determination of τ, τ_0, and $\alpha(\lambda)$ using the equations provided.

4 Execution

To set up the pulsed laser (purple light), as for the setup shown in 3, such that we get a ring-down-signal on the oscilloscope, an constant (not pulsed) adjustment Laser (red light) is used. Best case being that the adjustment Laser emits its light in the same direction relative to its mount as the pulsed Laser to its-.

Such the adjustment Laser can be setup relatively easy, because this light has a much higher and constant intensity than the pulsed Laser and has red light. Therefore the light emitted of the cavity is easily detectable and positionable in the middle of the output mirror with just a sheet of paper and adjustment of the cavitys (resonators) mirrors. Once done the adjustment Laser and the pulsed Laser is switched. The pulsed Laser is aligned with the optical axis of the setup. The detector and photo-multiplier are installed on the end of the resonator, so photons hitting the very small detector area are detected, that is next to very much noise. The pulsed Laser (pL) does not hit the detector, so the mirrors of the cavity get adjusted until we get the ring-down signal in sufficient intensity, meaning we the beam is directed directly on the detectors area. There is a normed systematically iterative pattern for getting the pulsed signals hitting the detectors area. The screws responsible for the position on the horizontal and vertical axis on each mirror are adjusted, such that the noise signal shows a maximum. Maximum noise occurs in close range around the detector. One mirror at a time is set. After, the arithmetic mean of the equivalent axis values (horizontal A and horizontal B and analogous) of the two mirrors is calculated and set as new value for both screws. This means taking two points close to the detector, then taking the point in the middle of their connecting straight as a new starting point for then finding higher local-noise-maxima and asap the detectors surface.
Once the setup is complete, the measurements of the incoming ring-down-signals are taken in two scattering media in the cavity of the resonator. One is Air of atmospheric pressure and the other one is in a vacuum. Such two and a half pairs of values get taken. For an sufficient amount of values to get a reliable average, it should be more values in relevance, but the signal was lost again and could not in time be found again while adjusting the mirrors to the new refractive index. Because the measurements are recorded alternately in air and vacuum and the signal takes so long to be found again once lost (even with the average of measurements method as explained before), it is necessary to set the new medium successively by getting a bit of air in or out and then setting the mirrors so the beam hits the detector optimal until the wanted pressure is reached.

5 Results

First, the series of measurements of the ring-down-signals were recorded. Five values should be recorded for air and vacuum in order to achieve a statistically stable result. However, this failed and only four values for air and 3 values for vacuum could be recorded.
An example of the measurement data can be seen in Fig. (2), where only the area within the two vertical red lines is relevant for determining the scattering coefficient.

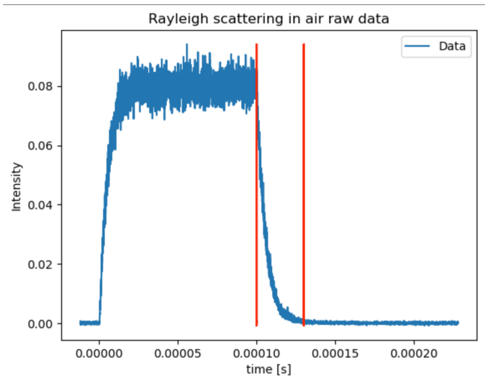

Figure 2: Plot of the measurement data for Rayleigh scattering in air, with the relevant area (intensity drop) marked by two red lines

The marked area was now fitted with the curvefit module from Scipy (The plot for the measurement data from Fig.(2) is shown in Fig.(3)).

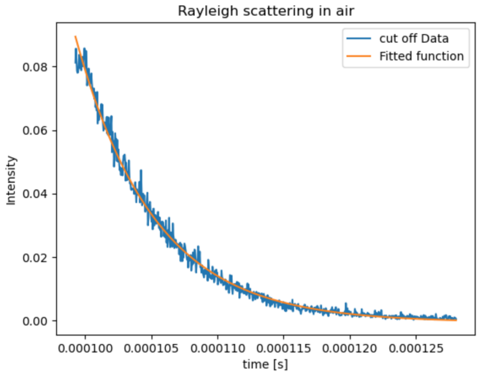

Figure 3: Plot and fit of the relevant area in Fig.(2)

The data were fitted with an exponential function of the following form:

$$f(x) = A \cdot e^{bx} + C \tag{1}$$

This procedure was repeated with all series of measurements, whereby the relevant parameters of the fit are only the exponents b, as these correspond to the inverse ring down times.

All fit parameters are listed with their errors in Tab.(1):

Table 1: List of the fit parameters b (from Eq.()) of the respective fits of the measurement data, where the values b_i in $[\frac{10^5}{s}]$ and Δb_i in $[\frac{10^3}{s}]$ are given

	b_1	Δb_1	b_2	Δb_2	b_3	Δb_3	b_4	Δb_4
Air	-1.69	1.26	-1.71	1.38	-2.04	1.86	-2.25	1.98
Vacuum	-1.11	1.01	-1.50	1.20	-1.62	1.36		

These values can now be converted into the respective τ value (shown in Tab.(2)):

$$b_i = -\frac{1}{\tau_i} \tag{2}$$

where the error is calculated using Gauss:

$$\Delta\tau_i = \sqrt{\left(\frac{\partial\tau_i}{\partial b_i} \cdot \Delta b_i\right)^2} \tag{3}$$

$$\Delta\tau_i = \frac{\Delta b_i}{b_i^2} \tag{4}$$

Table 2: Converted values from Tab.(1), where the values τ_i are given in $10^{-6}s]$ and $\Delta\tau_i$ are given in $[10^{-8}s]$

	τ_1	$\Delta\tau_1$	τ_2	$\Delta\tau_2$	τ_3	$\Delta\tau_3$	τ_4	$\Delta\tau_4$
Air	5.92	4.41	5.85	4.72	4.90	4.48	4.43	3.89
Vacuum	9.01	8.23	6.67	5.32	6.16	5.15		

Now the scattering coefficient can be calculated using Eq.(4), whereby the mean values of the values from Tab.(2) have been used for the values of τ_{air} and τ_{vac}:

$$\beta = \frac{1}{c}\left(\frac{1}{<\tau_{air}>} - \frac{1}{<\tau_{vac}>}\right) \tag{5}$$

$$\beta = 1.74 \cdot 10^{-4} \pm 1.29 \cdot 10^{-7}[m^{-1}] \tag{6}$$

where the mean values are:

$$<\tau_{air}> = 5.27 \cdot 10^{-6} \pm 2.19 \cdot 10^{-8}[s] \tag{7}$$

$$<\tau_{vac}> = 7.28 \cdot 10^{-6} 3.69 \cdot 10^{-8}[s] \tag{8}$$

The error from the mean values was determined using Gaus:

$$\Delta <x> = \frac{1}{n}\sqrt{\sum \Delta x_i^2} \tag{9}$$

The error of β was determined as follows:

$$\Delta\beta = \sqrt{\left(\frac{\Delta\tau_{air}}{\tau_{air}^2}\right)^2 + \left(\frac{\Delta\tau_{vac}}{\tau_{vac}^2}\right)^2} \tag{10}$$

6 Discussion

For the scattering coefficient in this Experiment following value was aquired:

$$\beta = 1.74 \cdot 10^{-4} \pm 1.29 \cdot 10^{-7} [m^{-1}] \tag{11}$$

Comparing this to the literature value of $\beta = 4.19 \cdot 10^{-5}$ one can see that this value is not within three times the error interval of our result.
This is probably due to the large fluctuations in the recorded values for the ring down time τ. Because few measurements were recorded on the day of the experiment, no statically stable result could be achieved.
In addition, some difficulties were observed in adjusting the mirrors. Some of these were not as sensitive as they should be.
Despite all this, the experiment can be considered a success, although the setup should be adjusted.

References

[1] https://wiki.physik.fu-berlin.de/fp/_media/private:versuchsanleitung.pdf