

# RAYLEIGH SCATTERING THERMOMETRY AT LOW TEMPERATURE USING NITROGEN AND CARBON DIOXIDE WITH HIGH RAYLEIGH SCATTERING CROSS SECTION AREA

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**Abstract:** Rayleigh scattering thermometry is a practically applicable non-invasive measurement technique to investigate a gas flow field. Experimental results of Rayleigh scattering thermometry at the low temperature region from 230 K to 310 K are discussed with applying nitrogen and carbon dioxide. More precise measurement can be achieved by applying the carbon dioxide gas with high Rayleigh scattering cross section area. Several issues which should be handled importantly for a closed visualization experiment system at low temperature, such as frost on windows, sealing for insulation, thermal stress-induced polarization, and visualization set-up, are discussed.

**Keywords:** Rayleigh scattering thermometry, flow visualization, cryogenic temperature, low temperature

## 1. INTRODUCTION

In the extreme flow field conditions, such as flame, combustion, and plasmas, non-invasive thermometry using laser is free to installation of temperature sensors, disturbance in the fluid flow, extremely high temperature condition, chemically reactive situations [1]. Since most of non-invasive thermometry techniques are based on the application of laser induced electromagnetic spectrum, it can be extended to 2-dimensional diagnostic and its visualized distribution of temperature gives enormous information about a flow field. Although flow visualization method has been widely utilized to study flame or combustion fields, there has been relatively little research regarding non-invasive thermometry at low temperature. In the experimental work at low temperature, there are various technical problems for detecting temperature: installation of feedthrough passing through the vacuum insulation or test chamber, thermal contact between of a sensor and an object, thermal conduction through a current lead, and difficulty of removal and adhesion of sensor on an object. Accordingly, non-invasive measurement method is very useful to measure the temperature and visualize a complex and dynamic flow field at low temperature. Some researchers tried to measure low temperature with acetone laser induced fluorescence (LIF) at low temperature [2, 3]. Although LIF thermometry gives the largest scattering signal among temperature diagnostic techniques based on scattering, there is the limitation of temperature decrease due to a condensation problem of seeder, such as acetone, biacetyl, or acetaldehyde. Though there are some gases with low condensation temperature, the gases are hazardous to human body. OH

planar-LIF (PLIF) and Raman imaging for 2-dimensional visualization were conducted to investigate cryogenic jet flame [4, 5]. For open flow field like a flame field, the problems of thermal insulation and a closed system for low temperature was not considered. Besides, OH PLIF is only able to diagnose a flame field due to the requirement of OH seeder produced in a combustion process. Raman scattering cross section area is approximately 100 to 1000 times smaller than Rayleigh scattering cross section area [6]. Since Rayleigh scattering thermometry does not require seed material, the problems regarding to it, such as uniformly seeding of high speed gases, corrosion, and pollution problem, are not considered and practical application can be easily utilized [7]. Rayleigh scattering thermometry was applied to diagnose the temperature field of a pulse tube cryocooler in the range until 200 K [8]. However, the calibration process before diagnostic was carried out only until 270 K and the quartz-glass pulse tube part was not thermally insulated. A cryostat and optical window application for laser passage and observation, and calibration step are needed to visualize the internal temperature field of a cryocooler or a cryogenic liquid transfer system with stable and reasonable condition. This paper presents the experimental approach of 1-D calibration of Rayleigh scattering thermometry at low temperature and experimental results. Some experimental modifications, like using the gas with high scattering cross section area, are conducted to obtain the precise measurement of scattering signal. Especially, we discuss some experimental problems caused by low temperature environment and suggest its solution.

## 2. RAYLEIGH SCATTERING THERMOMETRY

Rayleigh scattering intensity,  $I_s$ , is expressed as follows.

$$I_s = CIN\Omega l \left( \frac{d\sigma}{d\Omega} \right)_{\text{eff}} = \frac{PK}{T} \left( \frac{d\sigma}{d\Omega} \right)_{\text{eff}} \quad (1)$$

where  $C$  is a calibration constant of the collection optics,  $I$  is the incident laser intensity,  $N$  is the molecular number density,  $\Omega$  is the solid angle of the collection optics,  $\sigma$  is the Rayleigh scattering cross section area,  $l$  is the length of the laser beam segment imaged onto the detector. As shown in equation (1), Rayleigh scattering intensity is decided by various properties of optical system and gas. For

thermometry, the dependence between the scattering intensity and the molecular number density is only considered. By ideal gas equation of state, the molecular number density is converted to temperature and pressure of gas as the second term of equation (1), where  $P$  is gas pressure, and  $T$  is the gas temperature [9]. Also, since all the optical values are assumed as constant, these constants can be converted to the one representative constant,  $K$ . In equation (1), it is assumed that the all the values of optics are constant and the molecular number density of gas is simply decided by ideal gas equation of state. Rayleigh scattering cross section area is inversely proportional to the wavelength of incident laser beam [9]. Besides, ultraviolet (UV) laser gives the reduced reflection from the surfaces or from particles within the laser sheet and increases Rayleigh cross section area of gas [10]. Many recent researches apply deep UV laser to Rayleigh scattering method and we also apply 266 nm ultraviolet laser to this research. As shown in equation (1), when the temperature is decreased, the molecular density is increased and scattering intensity is also increased. For low temperature measurement, Rayleigh scattering thermometry is a suitable method to obtain high signal-to-noise ratio which means high-quality accuracy and resolution in thermometry. Constant  $K$  in equation (1) can be evaluated by temperature, pressure, and scattering intensity which is measured at reference condition. Therefore, temperature of gas is described as follows [11].

$$T = \frac{I_{ref} P}{I_s P_{ref}} T_{ref} \quad (2)$$

Since the change of Rayleigh scattering cross section area is negligible as pressure and temperature change and the used gas is not a mixture, differential Rayleigh scattering cross section area term in equation (1) is also constant and is removed as shown in equation (2).

### 3. EXPERIMENTAL SET-UP

As shown in Fig. 1, the 266 nm laser beam is formed from 10 Hz pulsed Nd: YAG laser source (Continuum, Powerlite Precision II 8000) with fourth harmonic generator which generates horizontally polarized 266 nm laser from 1064 nm laser. Laser beam is reflected at Nd:YAG laser line mirror for 266 nm and passes through the iris to remove the edge unrequired part of laser beam and to reduce the beam size to eliminate back-reflection light in the test cell. The test cell is rectangular shape and extended adaptor parts are installed at both side of laser inlet and outlet to reduce the back reflection inside test cell. Test cell is made of copper to make thermal equilibrium state easily in the cell. Laser inlet, outlet, and observation window of the test cell are UV fused silica window (Thorlab, high-precision laser window) with 1 inch diameter and 5 mm thickness. These windows are sealed with indium wire gasket for cryogenic sealing [12]. 5 E-type thermocouples are installed crosswise vertically in the center of the test cell and located on the rear position of laser beam path line. 2 silicon diode sensors for cryogenic temperature measurement are installed on outside surface of

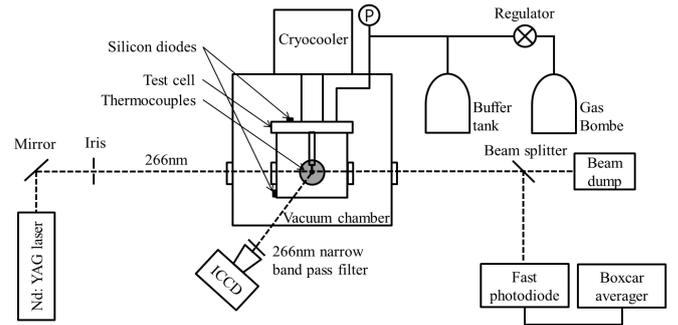


Figure 1. Schematic diagram of experimental apparatus

the test cell to check the degree of thermal equilibrium between the test cell and the charged gas. Pressure in the test cell is measured from pressure transducer (Honeywell, 1000 kPa, 0.1% acc.). Rayleigh scattering makes scattering light with the same wavelength of an incident laser beam due to elastic scattering characteristic, and the reflected background light from windows and walls also have the same wavelength with it. Accordingly, it is not possible to capture an interesting scattering signal individually from the background signal. The reflected background signal is reduced by a black mat finish of the test cell and using anti-reflection coated quartz window for 266 nm. Top part of the test cell is attached to the cold head of Stirling cryocooler (Cryotel CT, 4W cooling power @60 K) with cryogenic thermal grease (Apiezon N grease) for low temperature. Resistance type heater is installed around the cold head. To create efficient insulation and avoid frost or dew on the windows at low temperature, the test cell is located in a vacuum chamber, called a cryostat. The cryostat is made of 304 stainless steel and cylindrical shape with 188 mm diameter and 258 mm height. Degree of vacuum is maintained under 1 mTorr during experiment. The cryostat also has laser inlet, outlet, and observation windows, and these windows are sealed with rubber o-ring gasket and teflon. Laser inlet and observation window are 79.5 mm diameter and 5 mm thickness quartz. Outlet laser window of cryostat is 1 inch UV fused silica window and is installed on the adaptor with the inclined angle to prevent reflected laser beam from re-entering into the test cell. Additionally, to improve vacuum insulation and solve frost problem effectively, the copper panel coated with coconut charcoal which is a kind of moisture absorption material is installed around the test cell. Intensity of the Rayleigh scattering signal is measured by ICCD camera (Princeton Instruments, PI-MAX2). In front of ICCD camera, UV quartz lens (UKAoptics, 105 mm, f/4.0) and 266 nm narrow band pass filter (Edmund, CWL 266 nm, 55% transmission) is installed to collect only elastic Rayleigh scattering signal. Laser beam power is captured at photodiode (Thorlab DET25K, GaP detector) and boxcar averager (Stanford Research System, SR250 average) to compensate laser power fluctuation and drift. Before thermometry, the preliminary experiment using carbon dioxide is conducted to check the validation of experimental set-up by obtaining the linearity between Rayleigh scattering intensity and pressure at constant room temperature. As shown in Fig. 2, Rayleigh scattering intensity in which laser power fluctuation is

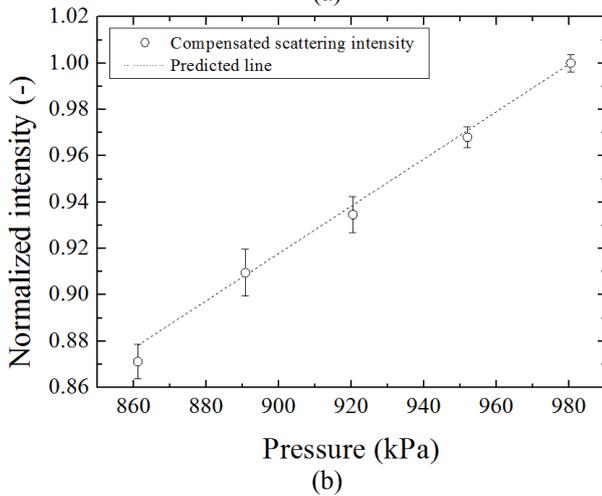
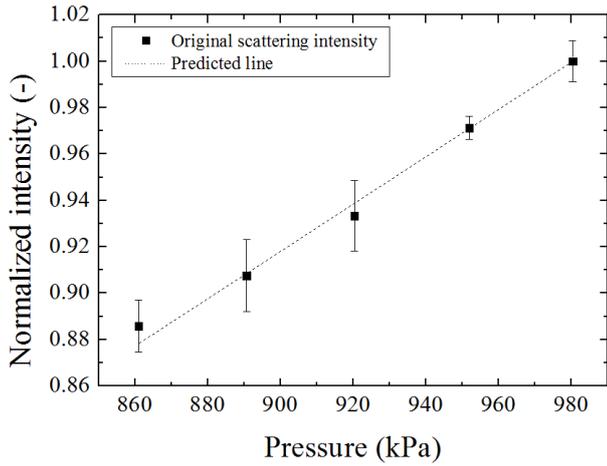


Figure 2. Linearity between pressure variation and (a) original Rayleigh scattering intensity and (b) compensated Rayleigh scattering intensity.

compensated by photodiode shows more precise value at each pressure point and is clearly proportional to the pressure of gas. We can achieve precision within 1% with 500 averaged-shots.

#### 4. EXPERIMENTAL RESULTS AND DISCUSSION

Test gases, nitrogen and carbon dioxide, are charged into the test cell and buffer tank. The inner volume of the test cell is 53 mL and the volume of the buffer tank which is maintained by normal temperature is 5 L which is sufficiently larger than the one of the test cell. Therefore, the pressure change is negligible during temperature change of charged gas. It is able to be assumed that the experiment is conducted under the constant pressure condition and the molecular density of the charged gas is only changed as temperature changes. In the experiment, the percentage of pressure change is maintained within 1% of initial pressure, 800 kPa. The initial temperature of the test cell and the charged gas is set as 230 K and 235 K for nitrogen and carbon dioxide, respectively. The variation of Rayleigh scattering intensity as temperature changes is shown in Fig. 3. Since the reference temperature is set as the initial low temperature, both of the measured normalized intensity and

the predicted line start at the value of one. The average error of the scattering measurement is 2.3 % in the case of nitrogen, and it is 1.2% in the case of carbon dioxide. Also, as shown in Fig. 3(a), the compensation of laser power fluctuation does not function effectively as measuring time is elapsed, approximately 1 hour, in the case of nitrogen. Applying horizontally polarized laser, Rayleigh cross section area of carbon dioxide is 10.7 times greater than the one of nitrogen [13]. In this experiment, for carbon dioxide, it is measured as 13.5 times greater than the one of nitrogen. Therefore, the preferable measurement with high precision is achieved in applying carbon dioxide with high Rayleigh scattering cross section area.

The important feature of these experimental results is significant distinction between the predicted line and measured scattering intensity. The scattering intensity is remarkably decreased by approximately one-fifth and one-third for carbon dioxide and nitrogen respectively as temperature is increased. On the other hand, the scattering intensity is increased more than expected as temperature is decreased when the reference value is set as 290 K. The scattering intensity is rapidly decreased until 270 K and the slope of intensity curve comes to agree with the predicted line after 270 K. Since the predicted line is decided by the gas properties which are measured during experiment, this

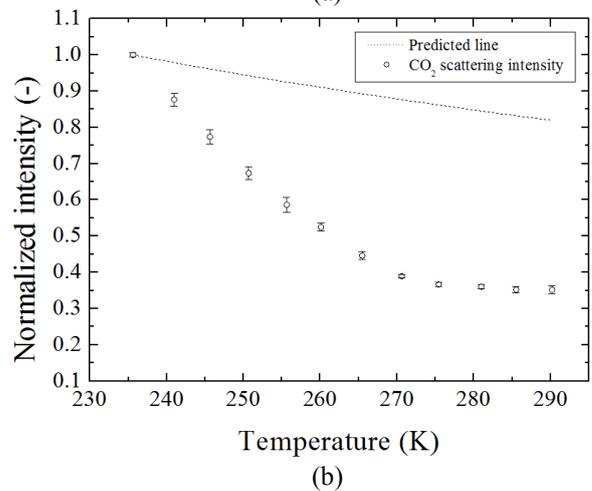
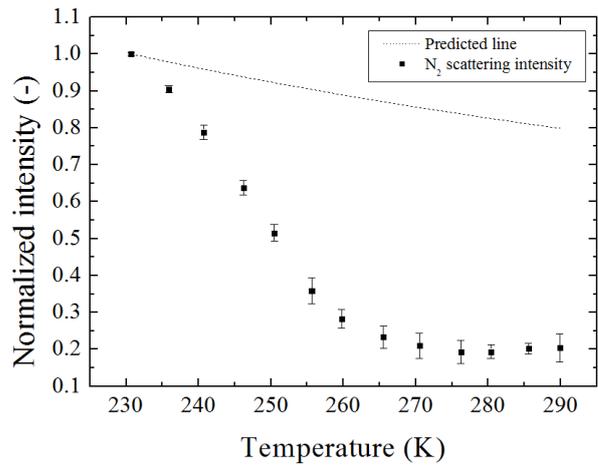


Figure 3. Rayleigh scattering intensity variation with temperature change: (a) nitrogen and (b) carbon dioxide

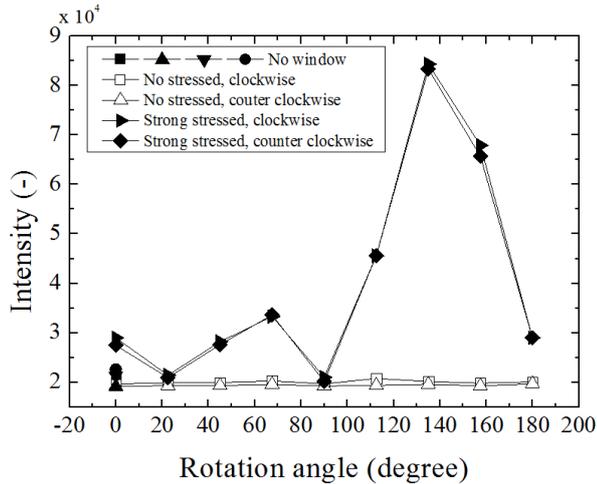


Figure 4. The mechanical stress effect of laser window on scattering intensity

phenomenon is independent of the gas properties and is caused by the temperature change on the laser window. In previous researches, there are thermal or mechanical stress-induced polarization effects of windows [14-16]. The simple experiment is conducted to check the stress effect on the polarization of laser window and the results is shown in Fig. 4. The same product of the laser window which is installed as the laser inlet window of the test cell is used for this test. The polarization characteristic of the laser window can be evaluated by rotating it and simultaneously checking the scattering intensity because the scattering signal is changed with the polarization change of laser beam by rotation of the laser window. For this test, the scattering signal is not compensated by the photodiode signal to exclude the effect of beamsplitter. The laser window for the test is rotated with the same interval of angle clockwise and counter clockwise to check the polarization effect of the laser window and the hysteresis of it. The Rayleigh scattering signal without window at 0 degree point means the original scattering signal. The variation of scattering intensity applying no stressed window presents that it provides approximately the same scattering intensity of the condition without the laser window and the rotation of the laser window is irrelevant to the intensity. It means that there is no polarization effect of the no stressed laser window and it is the normal characteristic of an optic device, such as a window, a mirror, or a lens except for a polarizer or a wave plate (retarder). However, the stressed window tightened by screw bolts for vacuum sealing shows the major effect of rotation on the scattering intensity. It means that there is mechanical stress-induced polarization effect of the laser window when the mechanical stress for vacuum sealing joint to keep low temperature is applied. Despite the condition that extremely large stress is not applied, the stress effect on polarization is not negligible. As shown in Fig. 5, the photodiode signal, which collects the laser beam reflected from beamsplitter having different vertical and horizontal reflectance ratio, is increased when temperature is decreased. This result implies that there is the change of polarization effect in the laser window with temperature change. Using horizontally polarized laser, the laser gives zero or minimum scattering

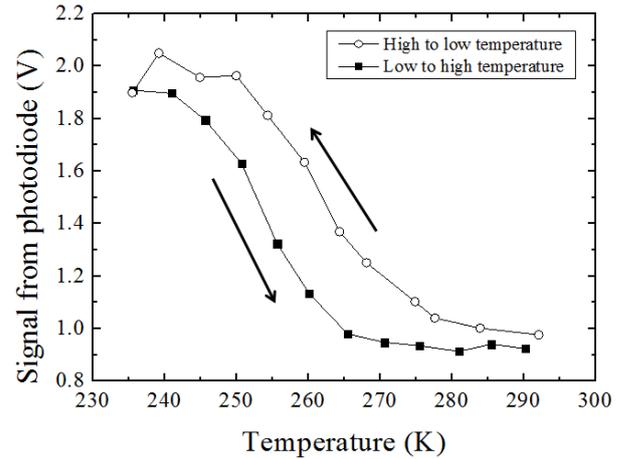


Figure 5. The variation of laser beam power reflected at beamsplitter with temperature change

signal among various linearly polarized laser [17]. The increase of scattering signal as temperature is decreased means the rise of vertical polarized component in the laser beam. According to these experimental results, it is reasonable that the significantly large scattering signal at low temperature is caused by increase of vertically polarization component in the laser beam because the laser passes through the laser window with thermally stress-induced polarization effect. Thermal stress is caused by different thermal expansion coefficients of quartz laser window, stainless steel bolts, and copper window cover and test cell.

As shown in Fig. 3, comparing the results of nitrogen and carbon dioxide, the thermal polarization effect at low temperature of mechanically stressed laser window occurs differently at each case. Also, as shown in Fig. 5, the polarization effect is different for the direction of temperature change. Accordingly, though it is needed to calibrate this polarization effect, but it is hard to achieve repeatability of thermal polarization effect experimentally in this experiment set-up. It is recommended that the laser windows are combined with the material of similar thermal expansion coefficient or the test cell is designed with no mechanical and thermal stress on glass like tube type glass application.

## 5. CONCLUSION

In this paper, 1-D Rayleigh scattering thermometry is conducted at low temperature with gas of high Rayleigh cross section area with horizontally polarized laser beam. The preferable measurement performance is achieved with using carbon dioxide of high Rayleigh cross section area. The problems of Rayleigh scattering experimental set-up for low temperature, such as window sealing, insulation, frost on windows, are solved. The thermal and mechanical stress-induced polarization effect is observed at the sealed and pressed laser window for sealing of the cryostat and the test cell. It is needed to consider the stress-free design of laser passing window for low temperature thermometry.

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