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COMPACT RAMAN LIDAR MEASUREMENT OF LIQUID AND VAPOR PHASE WATER UNDER THE INFLUENCE OF IONIZING RADIATION

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ABSTRACT
A compact Raman lidar has been developed for studying phase changes of water in the atmosphere under the influence of ionization radiation. The Raman lidar is operated at the wavelength of 349 nm and backscattered Raman signals of liquid and vapor phase water are detected at 396 and 400 nm, respectively. Alpha particles emitted from ^{241}Am of 9 MBq ionize air molecules in a scattering chamber, and the resulting ions lead to the formation of liquid water droplets. From the analysis of Raman signal intensities, it has been found that the increase in the liquid water Raman channel is approximately 3 times as much as the decrease in the vapor phase water Raman channel, which is consistent with the theoretical prediction based on the Raman cross-sections. In addition, the radius of the water droplet is estimated to be 0.2 μm.

1. INTRODUCTION
Monitoring of radioactive substances is indispensable for ensuring safety for the ongoing dismantling operation of the nuclear reactors damaged in the wake of the Great East Japan Earthquake. In addition to conventional proximity detectors such as a Geiger counter, stand-off detectors such as a gamma camera has been developed and applied at the site. For obtaining early warning, however, stand-off detection of aerosol particles that are produced under the influence of ionizing radiation is highly desirable.

In this study, we describe a compact Raman lidar system developed for studying phase changes of water in the atmosphere under the influence of ionization radiation. This is a modified version of the system originally developed for hydrogen leak detection: it has attained the detection of 1% hydrogen in the air with the stand-off distance of less than 50 m [1]. Radiation from radioactive materials ionizes molecules in the ambient atmosphere. The reaction of these ions or ion clusters with water vapor leads to the formation of hydrates. Overall, it is expected that water droplets are produced as a result of ionization. In this paper, the effect is studied in a chamber equipped with 9 MBq ^{241}Am sources that produce alpha particles (5.4 MeV). The signals observed for water vapor Raman and liquid water Raman channels are compared for monitoring the phase changes of water.

2. COMPACT RAMAN LIDAR
The compact Raman lidar has an in-line configuration, in which the same optics is employed as both transmitter and receiver. The 349 nm laser output (~100 μJ/pulse, 1 kHz) is generated as the third harmonic of a Nd:YLF laser. As shown in Fig. 1, the Raman backscattering signals of liquid water (396 nm) and water vapor (400 nm) are detected in addition to the hydrogen (380 nm) and nitrogen (408 nm) Raman signals. A filter wheel is used to switch the filters (3 nm FWHM) for 396 and 400 nm. The measurement is conducted with photon counting mode. A narrow field-of-view of 2 mrad is attained with the in-line optics. The maximum observation ranges for atmospheric nitrogen and water vapor detection are 70 and 50 m, respectively. The count of water vapor Raman channel is corrected for the contribution of liquid water Raman, which exhibits long-wavelength tail within the filter transmittance centered at 400 nm.

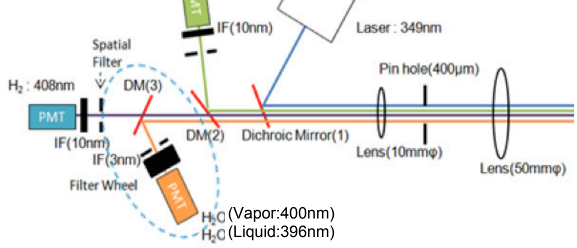

Fig. 1 Improved compact Raman lidar for water phase change observation.

3. RAMAN MEASUREMENT UNDER RADIATION

The radioactivity and radiation dose of the ^{241}Am source are 3 MBq and 50 mSv/h, respectively. Three pieces of sealed ^{241}Am source are employed for investigating the influence of radioactivity. Figure 2 shows the experimental setup of the radiation chamber, which can be humidified with wet towels soaked in a water-filled vessel. Under room temperature, the humidity inside the chamber reaches 90% after 30 - 60 min. The laser beam is passed through fused-silica glass windows, with its position located around 15 mm beneath the surface of the Am sources, in consideration of the relatively short range of alpha particles (a few cm).

4. RESULTS AND DISCUSSION

The experimental results obtained with the humidity of 90% are summarized in Fig. 3. Figure 3(a) and (b) show the Raman signal counts of liquid water and water vapor observed at 396 and 400 nm, respectively, after subtracting the background due to the atmospheric signals outside the chamber. As compared with the original condition in which all the Am sources were shielded, the liquid water counts increased by 16.8% whereas the water vapor counts decreased by 5.1% when all the Am sources were opened (9 MBq in total). The experiments were repeated under different levels of humidity with different numbers of Am sources opened.

If the increase of liquid water Raman counts was caused by water cluster, the decrease of water vapor Raman counts is considered as the reflection of the ionization effect, leading to the formation of water droplets. The Raman lidar signal expressed in terms of the number of photons can be written as

$$N(R) = n \cdot l \cdot s \cdot \frac{A_r}{R^2} \cdot P(R) \cdot f \,/\, \frac{hc}{\lambda}, \qquad (1)$$

where n: number density, s: differential cross-section of scattering, l: optical path length, A_r: receiver aperture, R: distance, $P(R)$: laser intensity, f: laser repetition frequency, λ: laser wavelength, h: Planck's coefficient, and c: speed of light. The differential cross-sections of liquid water and water vapor are 5.6×10^{-29} and 2.0×10^{-29} cm^2/sr, respectively [2-4]. Equation (1) indicates that the counted photon number is proportional to the value of ns. If we assume that the effective number densities of water vapor and liquid water molecules are the same, the net ionization effect as detected by the change in Raman signal intensities should be equal to the ratio of the differential cross-sections, which is calculated to be 2.8. This value is fairly close to the experimental result of the ratio of 3.3. Thus, the present results suggest that the changes observed in the two Raman channels are caused by the ionization process under the influence of alpha particles from the Am sources. Moreover, if we assume that each water cluster contributes to Raman scattering, the radius of water cluster is estimated to be 0.2 μm, a reasonable size for a water droplet.

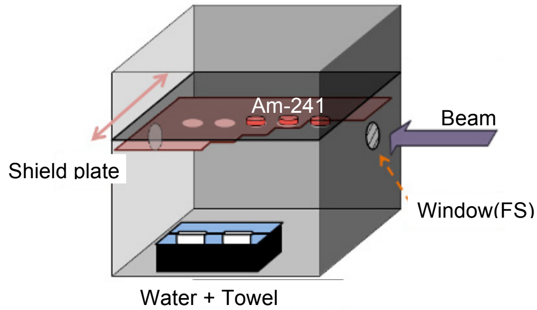

Fig. 2 Radiation chamber with humidifier.

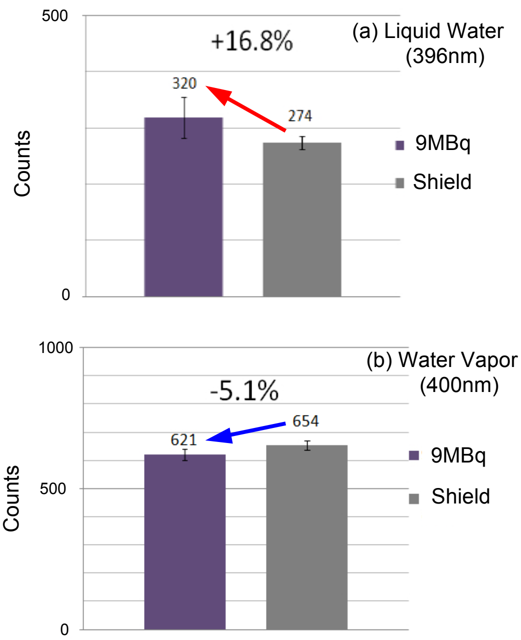

Fig. 3 Phase change measurement with 90% humidity.

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