Mechanism of sonoluminescence from K atom in aqueous solution
水溶液中 K 原子からのソノルミネセンス機構
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1. Introduction
Sonoluminescence (SL) can be used as a spectroscopic probe of species produced at bubble collapse. Studies of multibubble sonoluminescence (MBSL) from alkali-metal salt solutions have revealed emission from the excited alkali-metal atoms. However, there are several unsolved problems for the alkali-metal atom emission. The subject of the problem is where alkali-metal emission occurs, in gas phase or liquid phase. It is still under debate where alkali-metal emission occurs, how alkali-metal ions are reduced, and how alkali-metal atoms are excited. Choi et al. [1] investigated the concentration and acoustic power dependences of MBSL spectra from NaCl solution doped with ethanol. The close investigation of line width and intensity indicated that the sodium emission occurs in the gas phase. The purpose of the present paper is to investigate effects of saturation gas on line width and shift of alkali-metal emission. Potassium atom emission is favorable for evaluating the line-broadening, peak shift and symmetry since the two lines of K doublet are more widely separated than those of sodium doublet. We measured MBSL spectra from KCl aqueous solutions saturated with Ar, Xe and He gases at temperature of 15 °C at frequency of 148 kHz. The results indicate that the potassium emission occurs in gas phase within bubbles.

2. Experimental
We measured MBSL spectra in the 270 - 800 nm range from KCl aqueous solutions with concentration of 1 M. The ultrasonic frequency and power used are 148 kHz and 2.3 W, respectively, unless otherwise specified. A cylindrical cell was made of stainless steel. The size of the sample container was 46 mm in diameter and 150 mm in length. The top and bottom faces of the cell were equipped with a quartz glass window and a sandwich-type transducer, respectively. The temperature of the sample was controlled to 15 °C by circulating water. The solution was carefully degassed, and then re-gassed with rare gas for at least 2 hours. The signal from a function generator was amplified using a power amplifier and was matched for impedance to the transducer using a transformer. Emitted light was analyzed using a system of a monochromator and a cooled-CCD detector. Broad-band spectra were collected using a grating of 600 grooves/mm blazed at 300 nm. The narrow- band spectra around the K line were collected using a grating of 1200 grooves/mm blazed at 500 nm. The instrumental width was estimated to be 0.315 nm from the measurement of the He-Ne laser line. The spectral response was calibrated for detection efficiency against a standard halogen lamp and Xe lamp. The total power of the irradiated ultrasound was determined by calorimetry.

3. Results and discussion
Figures 1 (a)- 1(c) show the K atom emission from KCl solution saturated with Xe, Ar and He, respectively. A collecting time of the spectrum was 1, 3 and 30 min for the solution with Xe, Ar and He, respectively. A flame spectrum of KCl was also measured and indicated as a thin line in Fig.1. The doublet from the flame spectrum are at 766.5 nm (2P3/2→2S1/2) and 769.9 nm (2P1/2→2S1/2). For Xe and Ar-saturated KCl solutions, it is noticed that the spectral peaks are not shifted and broadened asymmetrically to the red side (longer wavelength) compared with the lines of the flame spectrum. The K line peaks in the case of Xe in Fig. 1(a) are prominent in comparison with that in the case of Ar in Fig. 1(b), and cannot be analyzed as single doublet. This result suggests that the spectrum of K atom emission is composed of two types of peaks, unshifted narrow line and shifted broad line. We also observed K atom emission from He-saturated solutions. On the contrary to the cases of Xe and Ar, the K lines for He broadened symmetrically and shifted to blue side by 0.21 nm.

As shown in Fig. 1, the line shape of K atom emission considerably depends on the kind of gas
saturated in the solution. The spectra from Xe and Ar saturated KCl solution suggest that the unshifted narrow line and shifted broadened line overlap each other. On the contrary, the unshifted narrow lines were not observed in the case of He and only shifted broadened line was obtained. Those results make us to expect that spectra of K atom in the case of Ar or Xe may be separated under certain experimental conditions. We performed a further experiment in Ar-saturated KCl solutions under various conditions by changing ultrasonic frequency and power. Eventually, we obtained a spectrum indicating clear separation of unshifted narrow line and shifted broad line at the frequency of 47.9 kHz and high acoustic power of 23 W, as shown in Fig. 2. The broadened lines shift to red side by about 0.6 nm.

The line broadening and shift shown in Figs. 1 and 2 can be explained in terms of potassium and rare gas interactions [2]. Studies of gas-phase spectroscopy have shown that the effect of He perturbers on the collisional broadening of K atom emission is markedly different from that of Xe and Ar perturbers. He perturbers cause slightly-asymmetric broadening toward the blue side, whereas Xe and Ar cause asymmetric broadening toward the red side. Similar results have been reported in the case of Na line [3]. We can estimate a relative gas density at bubble collapse from the peak shift and line width in the case of He by comparing with the spectroscopic data [2]. The comparison gives a relative density of 43.7 and 39.3 from the peak shift and line width, respectively. Our relative densities estimated are consistent with values reported by previous work [4]. Further analysis for the case of Ar and Xe are in progress.

4. Conclusion

Sonoluminescence from KCl aqueous solutions was observed at temperatures in the range of 15 - 40 °C at the frequency of 148 kHz for various rare-gases saturation. The line broadening of potassium atom emission was independent of amount of water vapor and final temperature at bubble collapse. MBSL spectra from Xe and Ar-saturated KCl solutions showed that the spectrum of potassium atom emission is composed of unshifted narrow line and shifted broad line. In He-saturated solutions, the K lines shifted to blue side, and broadened asymmetrically. The effects of dissolved gas are in good agreement with data by spectroscopic studies. Hence, we conclude that the site of alkali-metal emission is in gas phase inside bubbles. An origin of the unshifted narrow line is unclear, and it should be investigated in future.

References