Investigations on the local structure of liquid sulfur at photo-induced polymerization transition III

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It is well known that sulfur consists of S_8 ring molecules just above the melting point (112.8°C) and changes to be a mixture of long polymeric chains with 10^5 atoms and S_8 ring molecules at 159°C. We found from the laser-excited optical absorption measurements using a nanosecond pulsed laser and the specially designed quartz glass container, which stably maintains the thin liquid film less than 1μm, that the polymerization occurred by also the laser illumination [1, 2]. In addition, we found that there were intermediate products between S_8 rings and the polymeric chains, which were supposed to be the S_8 chains and the tadpole-shaped S_7-S molecules [2, 3]. Moreover, the ordered macroscopic molecules were observed by the strong pulsed laser illumination more than 60 mJ/cm^2 in the confined liquid sulfur film with the thickness less than a few micrometers [2, 4]. These results indicate that the molecular structure flexibly changes by the light illumination, and it is interesting to investigate how the local structure changes by the light illumination. Neutron diffraction is a powerful technique to clarify the local structure of liquid. Therefore, we performed the neutron diffraction measurement on HERMES.

To realize high temperature experiments more than 125°C and a light illumination, we use the SANS, WANS furnace used in the MLF [5]. Since enough scattering data of the liquid sample were not obtained in the previous experiments (21811, 22808), the inner diameter of the sample glass tube was changed from 6 mm to 10 mm, and the wavelength of the neutron beam was changed from 1.34 Å to 2.1964 Å. The incident beam intensity is approximately 5 times greater than the previous one. The white light from the mercury lamp (REX-250, Asahi Spectra) was used as the excitation light source.

Figure 1 shows the neutron diffraction patterns of liquid sulfur obtained by subtracting the data from the empty glass tube. The total data acquisition time for the light illumination was 7.17 hours (86 min x 5). The diffraction peak at 1.1 Å corresponds to the first peak observed by Winter *et al*. [6]. Since the peak increases above the polymerization temperature in the Winter's data, the distinct increase of the peak by the light illumination in the present study suggests that the polymeric chains are produced by the light illumination.

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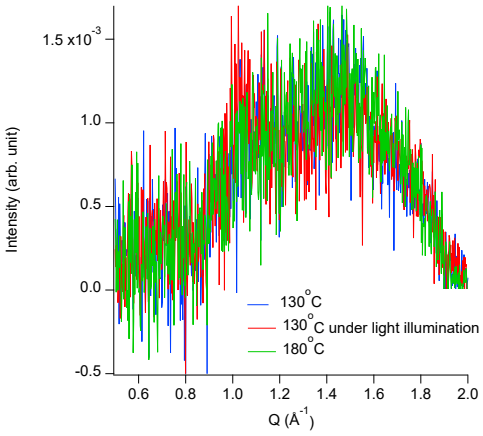

Fig. 1. Neutron diffraction pattern of liquid sulfur at 130°C without and with the light illumination, and at 180°C without the illumination.