MLF Experimental Report	提出日 Date of Report 2011/01/27
課題番号 Project No. 2010A0075 実験課題名 Title of experiment Relaxation mechanism in molecular liquid with strong intermolecular correlation 実験責任者名 Name of principal investigator Yukinobu Kawakita 所属 Affiliation JAEA/J-PARC	装置責任者 Name of responsible person Kenji Nakajima 装置名 Name of Instrument/(BL No.) AMATERAS(BL14) 実施日 Date of Experiment 2010/06/09～06/10 2010/11/24～11/26

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

Selenium Bromide (Se_2Br_2)

2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。)
Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

< Experimental method >

Quasi-elastic neutron scattering (QENS) of liquid Se2Br2 have been performed at three temperatures, 298 K, 373 K and 473K. Liquid sample sealed in SiO2 tube was put into an Aluminum container. High temperatures were achieved by using a cryo-furnace owned by the instrumental group. In addition to those measurements empty SiO2 glass tube was measured to be subtracted taking into account of absorption correction. The total measurement time was 34 hours for sample and 12 hours for empty cell and for instrument background with cryofurnace. Multi Ei method was applied so that incident neutron energies were 41.83, 7.72 and 3.13 meV.

< Results >

Chalcogen-halogen systems have a deep eutectic point around the equi-atomic composition in their phase diagrams, where the system is in the liquid state even at the room temperature. Selenium bromide consists of chain molecule with Br-Se-Se-Br form, where the bond angle and the dihedral angle are about 104° and 83° respectively [1]. Moreover, selenium bromide has strong inter-molecular correlation. As a result, each molecule doesn't have enough space to rotate freely due to molecular shape and high density.

2. 実験方法及び結果（つづき） Experimental method and results (continued)

Quasi-elastic neutron scattering (QENS) of liquid A_2X_2 (A=S, Se; X=Cl, Br) [2] and temperature dependence of Se_2Br_2 [3] suggested existence of two relaxation modes at low Q region. However, detailed Q dependence of QENS was not discussed because of short coverage of detectors of the spectrometer at that moment.

Fig 1 shows clear QENS signal in the scattering intensity with Ei=7.72meV, convenient to see Q region where inter-molecular correlations including orientational correlation between neighboring molecules are dominant. After subtraction of the cell contribution taking into account of absorption correction, fitting by model function was examined for spectra as a function as E at each Q point (Q-slice data). It is confirmed that sum of two Lorentzians, narrow and broad in E, can reproduce well them. Fig 2(a) shows the integral intensity of each component. Solid line shows the result of narrow component which corresponds to slow dynamics. The peak position shifts to lower Q direction and decrease intensity with increasing temperature. The broad component corresponding to fast dynamics also shifts to lower Q direction but its intensity becomes weaker with increasing temperature. Fig 2(b) shows the half width of half maximum (HWHM) of narrow component. It is natural that HWHM at room temperature is narrowest, but it is interesting that it exhibits similar value at 373 K and 473 K. This slow mode can be assigned as dynamical mode relating to molecular packing as function as density. On the other hand, the HWHM of broad component exhibit so called de Gennes narrowing as shown in Fig2(c). The Q value for narrowest HWHM shifts to lower Q direction with increasing temperature. Generally speaking, narrowing relates to strong correlation so that the fast mode relates to intermolecular correlation.

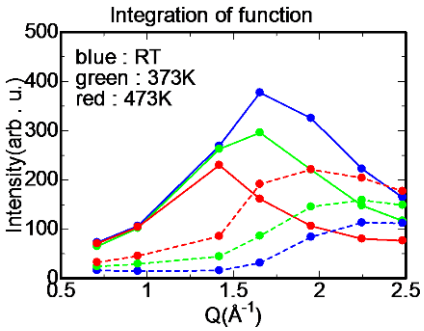

Fig2(a)

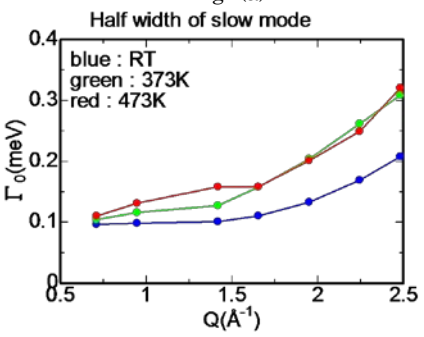

Fig2(b)

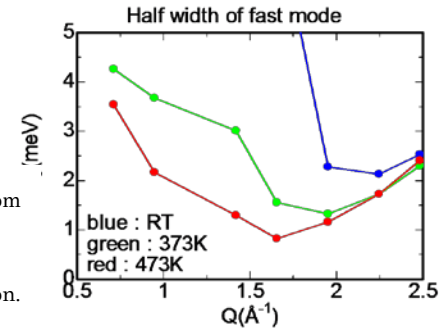

Fig2(c)

[1] K. Maruyama et al.; J. Phys. Soc,Jpn. 60 (1991) 3032

[2] M. Yao et al.; J. Phys. Soc. Jpn 66(1997)3115

[3] Y. Kawakita et al.; Physica B 385(2006)256

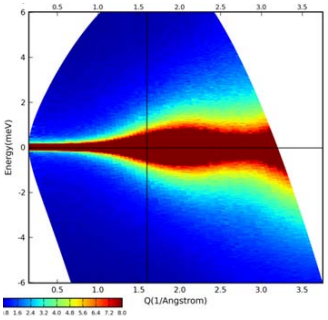

Fig1

Fig1. It shows the raw data at room temperature by using Ei=7.71.

Fig2. (a) shows integration of each function. Solid lines show slow mode's one and break line show fast mode's one. (b) show the HWHM of slow mode. (c) show the HWHM of fast mode.