MLF Experimental Report	提出日 Date of Report 2013/06/26
課題番号 Project No. 2012A0121 実験課題名 Title of experiment Intermolecular dynamics of strongly correlated molecular liquids of chalcogen-halogen mixtures 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 2012/04/12～04/14 2012/06/13～06/15

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)
Selenium Chloride (Se_2Cl_2)
Sulfur Chloride (S_2Cl_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 Se_2Cl_2, Se_2Br_2 and S_2Cl_2 were performed at room temperature. To reduce total dynamical structure factors, sample container and instrumental background were carefully measured. In order to obtain real space and real time information, so called van Hove function, $G(r,t)$, both measurements over a wide momentum-transfer (Q) range with moderate energy resolution and in a low Q region with high energy-resolution are necessary. Then multi E_i method with incident neutron energies of 93.83, 23.65, 10.51 meV was applied.

< Results >

Chalcogen-halogen systems have a deep eutectic point around the equi-atomic composition in their phase diagrams whose melting points are below room temperature. These eutectic molecular liquids consist of X-A-A-X chain molecules (A= S, Se; X=Cl, Br). In the case of Se_2Cl_2, EXAFS and neutron diffraction measurements suggested that this molecule has a gauche shape with the bond angle of 102° and the dihedral angle of 85° and that strong orientational correlation between neighboring molecules exists [1].

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

The Gauche molecular shape and relatively high number density prevent molecules to rotate freely without any interference from the neighbors. As a result, inter-molecular correlation is overlapped with intra-molecular one in the total pair-distribution functions. S_2Cl_2 is a little smaller molecule than Se_2Cl_2, and then dissolution between intra- and inter-molecular correlations in the pair distribution function g(r) is relatively better.

Quasi-elastic neutron scattering (QENS) of liquid A_2X_2 (A=S, Se; X=Cl, Br) [2] and temperature dependence of QENS for Se_2Br_2 [3] suggested that two relaxation modes, fast and slow modes, exist at low Q region and the fast mode relates to dynamics of neighboring molecules and the slow mode relates to translational diffusion [4].

Figure 1 shows the scattering intensity of liquid S_2Cl_2 in a quartz cell with E_i = 10.51 meV. Clear quasi-elastic spectrum was obtained where two relaxation modes are distinguishable. The structure factors obtained by integration of dynamical structure factor $S(Q,E)$ with E_i = 93.8 meV in the E range from -20 meV to 20 meV are shown in Fig. 2. Although the structure factors still include the scattering contribution from the quartz cell, the obtained I(Q)s are consistent with results from static structure studies. The Q dependences of quasi-elastic scattering are shown in Fig. 3. Real space and real time analyses for these spectra are now undergoing.

[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

[4] H. Shimakura et al; J. Phys.: Conf. Ser. 340(2012)012080

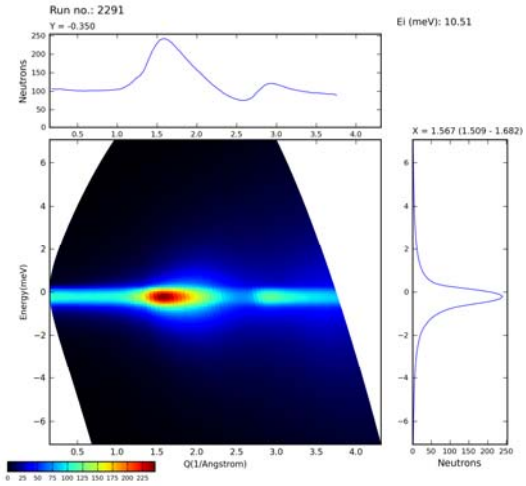

Fig. 1 Scattering intensity $I(Q,E)$ of liquid S_2Cl_2 in a quartz cell at room temperature.

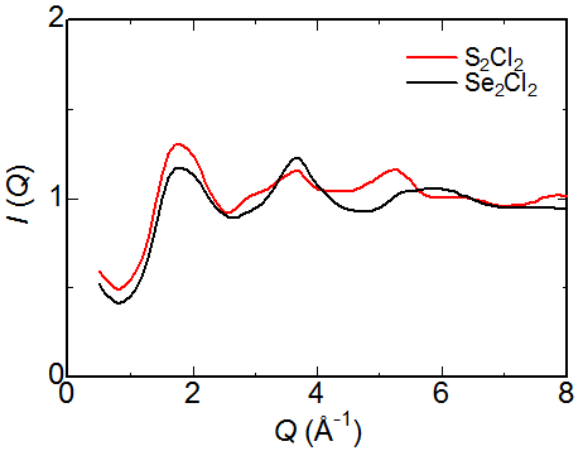

Fig. 2 Structure factors of liquid S_2Cl_2 and Se_2Cl_2 obtained by E-integration of $S(Q,E)$.

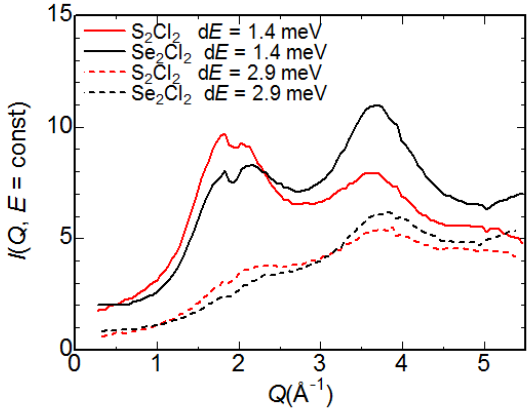

Fig. 3 Q-dependence of dynamic structure factors of liquid S_2Cl_2 and Se_2Cl_2 at E=1.4 and 2.9 meV.