（ ※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。）

CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 2017/6/21 承認者 Approver jun-ichi Suzuki 提出日 Date of Report 2017/5/16
課題番号 Project No. 2015A0087 実験課題名 Title of experiment Small-angle neutron scattering of liquid Au-Si – investigation on micro-inhomogeneity in homogeneous liquid in deep eutectic system 実験責任者名 Name of principal investigator Yoshifumi Sakaguchi 所属 Affiliation CROSS	装置責任者 Name of Instrument scientist Jun-ichi Suzuki 装置名 Name of Instrument/（BL No.） TAIKAN (BL15) 実施日 Date of Experiment December 15 9:00 –17 9:00, 2016

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.

$Au_{81.4}Si_{18.6}$ (liquid in the quartz glass cell) (eutectic composition)

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:

Fig.1 shows the phase diagram of Au-Si system. In this experiment, we measure small-angle neutron scattering (SANS) in the liquid phase of the eutectic composition, $Au_{81.4}Si_{18.6}$. To measure the SANS of the liquid phase, we need a furnace and a sample container. Fig.2 show the high temperature furnace, which was specially designed for SANS. Fig. 3 shows the quartz glass cell with $Au_{81.4}Si_{18.6}$ (0.5mm thickness) in an alumina tube. Such thin sample thickness was required because of the large absorption cross-section of gold. Beside the thickness, it is required to consider about “sealing” the liquid sample in a container because gold is easy to be radio-activated. Usually, such a glass cell is broken when the sample solidifies due to an abrupt volume change at the solidification.

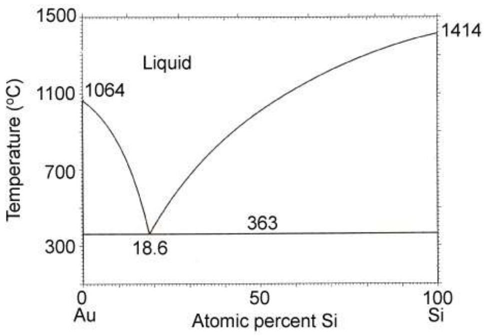

Fig.1


Fig. 2


Fig. 3

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

Therefore, the sample was prepared as small as possible to just fill in the thin samples space with the thickness of 0.5 mm with a diameter of 8 mm. Also, the glass cell with the $Au_{81.4}Si_{18.6}$ sample was sealed in a larger quartz glass tube again to prevent a leakage of the vapor when the glass cell is broken. The holes in the alumina tube do not interrupt both the incident neutron beam and the scattered beam. The aluminum windows with the thickness of 0.5mm minimize the absorption on the neutron beam paths. Prior to the SANS measurement of the sample, we measured the empty cell. The SANS measurements of the sample were performed from room temperature to 800°C. The proton beam power in the measurements was 150kW.

Experimental results:

Fig. 4 shows the SANS profiles of crystalline (room temperature) and liquid (400, 600, and 800°C) $Au_{81.4}Si_{18.6}$. At room temperature, the alloy is composed of separated two phases, Au and Si. The separated two phases form "eutectic structure", in which two types of lamellars alternatively align with the inter-lamellar spacing of several micrometers. The large SANS curve, which follows approximately Q^{-4}, is considered to show a sharp interface between the two phases. This large SANS intensity eventually decreases when the alloy melts by heating (Fig. 5). Usually, it is expected that such a SANS intensity completely disappears due to a formation of "homogeneous" liquid, which means that there is no order with nanometer-scale. However, an apparent SANS intensity is observed in the liquid state in the *Q*-range less than 0.02 Å^{-1}. Also, there could be a small hump (or shoulder) around 0.1 Å^{-1}, although the influence of the analytical procedure and the data processing should be carefully considered. As of now, it is not easy to conclude the physical origin of the SANS intensity in the liquid state. One possibility is the compositional fluctuation, which is generally observed around "critical point" of a phase separation in liquid. In this case, the SANS intensity follows so-called Orenstein-Zernike relationship, $I(Q) = I(0) / (1+\zeta^2 Q^2)$, where ζ is the correlation length, in the *Q*-range, usually from 0.05 to 0.24 Å^{-1}. Itami *et al.* pointed out a presence of such type of compositional fluctuation to explain the anomalous behavior in their experimental result of the electrical resistivity measurement [1], and asserted that the extremely low melting point at the eutectic composition is related to the presence of the composition fluctuation. However, we observe much larger SANS intensity below Q= 0.03 Å^{-1}, which is insensitive to the temperature variation. Probably, there could be other physical origin to explain about the large SANS intensity.

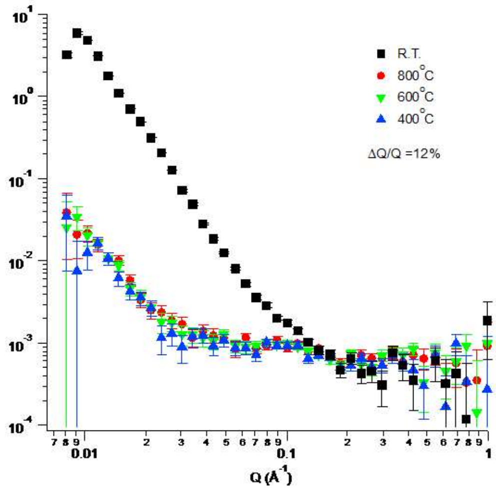

Fig.4

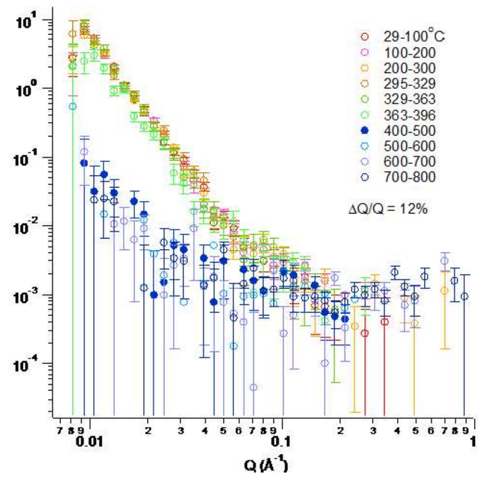

Fig. 5

[1] T. Itami *et al.*, J. Non-Cryst. Solids 353 (2007) 3011.