



	<b>Experiment title:</b> Partial structure factors of liquid amalgams: Existence of chemical order in liquid states	<b>Experiment number:</b> SC-500
<b>Beamline:</b> BM02	<b>Date of experiment:</b> from: 26 Nov. 1998 to: 30 Nov. 1998	<b>Date of report:</b> 27 August 1999
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### Report:

Since the resistivity measurements of liquid (l-) Na-Hg alloys by Müller in 1910, much interest has been devoted to alkali metal-Hg amalgams. The electronic properties such as resistivity, thermopower, and magnetic susceptibility and the thermodynamic properties such as density and electromotive force show unusual behaviour, and therefore, these alloys have been believed to form a kind of chemical order in the liquid states [1]. Although structural information is crucial to clarify this speculation and for further discussions, only total structure factors of l-Na-Hg alloys near the melting points were available until now. Anomalous x-ray scattering is a still developing technique that allows us to obtain information on the local structures of each constituent element.

We have carried out the experiment for l-Rb<sub>60</sub>Hg<sub>40</sub> at 200 and 500 °C using a 7-circle spectrometer at BM02. The energies of the incident x-rays were chosen at 5 and 287 eV below the Hg L<sub>III</sub> (12286 eV) and 5 and 127 eV below the Rb K (15202 eV) edges. The scattering x-ray photons were energy-analysed using a flat monochromator of pyro-graphite, and acquired using a photon counter. High temperatures were achieved using an Mo resistant heater in an internally heated vessel filled with He gas, having Kapton windows covering a scattering angle range of 0-90°. The l-Rb<sub>60</sub>Hg<sub>40</sub> sample was contained in a single-crystal sapphire cell with a wall thickness of 250 µm and a sample thickness of about 10 µm, which is almost transparent to x-rays and resistant against the chemical corrosion by the hot sample. The scattering contributions by the sample cell, He gas, and the Kapton windows were

subtracted using scattering data from the empty cell measurement in the same geometry. The x-ray absorption by the sample was estimated using theoretical values.

In the figure below, full circles show the total scattering intensities,  $I(Q)$ , measured at 127 eV below the Rb K edge, which has almost no anomalous dispersion effect, evaluated after the above data corrections. From the first peak around  $2 \text{ \AA}^{-1}$  to higher  $Q$ , oscillations seem to be those typical of normal liquid metals. In the  $Q$  region below  $1.5 \text{ \AA}^{-1}$ , however, two characteristic features appear in the spectrum, i.e., a prepeak around  $1.2 \text{ \AA}^{-1}$  and a strong scattering enhancement at small  $Q$  region. These features, the prepeak and the enhancement, suggest an intermediate-range structural order with the correlation length of about  $5.3 \text{ \AA}$  and a strong density and/or concentration fluctuation in the liquid, respectively. We calculated the difference between the scattering intensities near and far from the Rb K and the Hg  $L_{III}$  absorption edges,  $\Delta_{Rb}I(Q)$  and  $\Delta_{Hg}I(Q)$ .  $\Delta_{Rb}I(Q)$  has no prepeak and no enhancement in the low  $Q$  region, while  $\Delta_{Hg}I(Q)$  shows much stronger enhancement of the scattering intensity than that in  $I(Q)$ . Each scattering-intensity consists of three partial structure factors of Rb-Rb, Rb-Hg, and Hg-Hg correlations in the form of linear combination. The weighting factors at  $Q = 2 \text{ \AA}^{-1}$  are indicated in the table, and change very slowly with  $Q$ . It should be noted that the Hg-Hg correlation has a small negative contribution in  $\Delta_{Rb}I(Q)$ , whereas it has much larger contribution in  $\Delta_{Hg}I(Q)$  than that in  $I(Q)$ . It is plausible to speculate that the intermediate-range structural order and a strong density and/or concentration fluctuation in  $1\text{-Rb}_{60}\text{Hg}_{40}$  happen only in the Hg-Hg partial structure.

Further discussions for the evaluation of the differential structure factors or the partial structure factors are now in progress [2]. Developments of the experimental technique, such as an improvement of the detector system and simultaneous x-ray absorption measurements, are essential for the precise analyses of the anomalous x-ray scattering data.

[1] for example, M. Shimoji, *Liquid Metals*, (Academic Press, London, 1977).

[2] S. Hosokawa, A. Goldbach, W.-C. Pilgrim, J.-F. Bérar and D. Raoux, *Chem. Phys. Lett.*, in preparation.

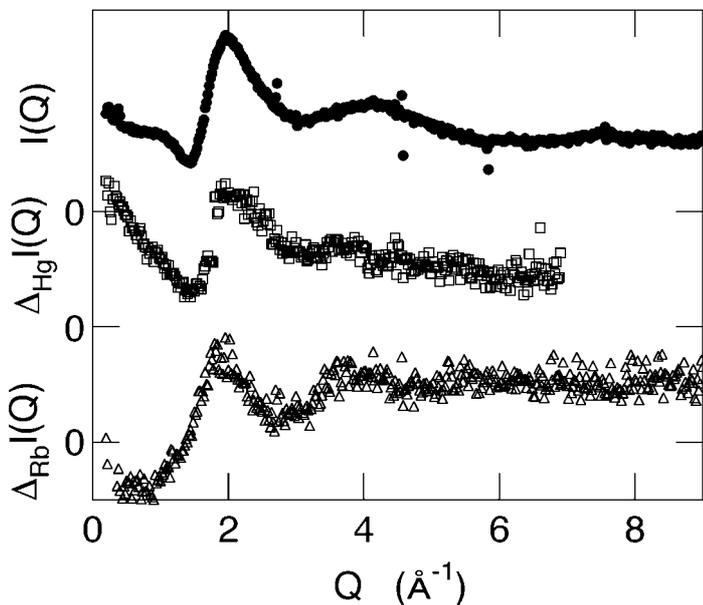


Table. Weighting factors of partial structure factors in  $I(Q)$ ,  $\Delta_{Rb}I(Q)$ , and  $\Delta_{Hg}I(Q)$  at  $Q = 2 \text{ \AA}^{-1}$ .

	$I(Q)$	$\Delta_{Rb}I(Q)$	$\Delta_{Hg}I(Q)$
Rb-Rb	0.158	0.448	0.005
Rb-Hg	0.479	0.708	0.447
Hg-Hg	0.363	<u>-0.156</u>	<u>0.548</u>

Figure. Differences of the scattering intensities close to the Rb K and the Hg  $L_{III}$  absorption edges, together with the total scattering intensity at 127 eV below the Rb K edge.