X-ray Diffraction Measurements for Expanded Fluid Mercury using Synchrotron Radiation

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1.Introduction

Liquid Hg is transformed to an insulating state when it is expanded to liquid-vapour critical point (crtical data of Hg [1]: Tc= 1470°C, pc= 1673 bar, dc= 5.8 gcm^{-3}). Many investigations have been made over the last three decades, focused on the metal-nonmetal (M-NM) transition in fluid Hg. It was found that physical properties such as electrical conductivity, thermopower, Hall coefficient, potical reflectivity, NMR sound velocity and the equation of state for fluid Hg are substantially changed at the density of about 9 gcm^{-3} where the M-NM transition starts to occur.

To understand the M-NM transision it is very important to investigate how the atomic arrangement is changed when liquid Hg is expanded. Recently x-ray diffraction measurements using an in-house x- ray source were made for expanded fluid Hg, giving information on the first neigh-bour coordination [2]. In the present paper we report for the first time the new results of the x-ray diffraction measurements using synchrotron radiation, extending from the liquid to the dense vapour region, which enable us to get more detailed information about the atomic configuration.

2.Experimental

We have performed energy- dispersive x-ray diffraction measurements for expanded fluid Hg on the BL-04B1 at Spring-8. The storage ring was operated at 8 GeV with 20mA during the present experiment. White x-rays were generated through the bending magnet ranging energy up to 150 KeV. The size of x-ray beam was made smaller down to 0.2 × 0.2 mm^2 using the horizontal and vertical tungsten slits located at the upper stream. The beam was directly introduced into the high pressure vessel through the Be window of the vessel. As a result the background noise due to the secondary x-rays in the hutch was substantially reduced.
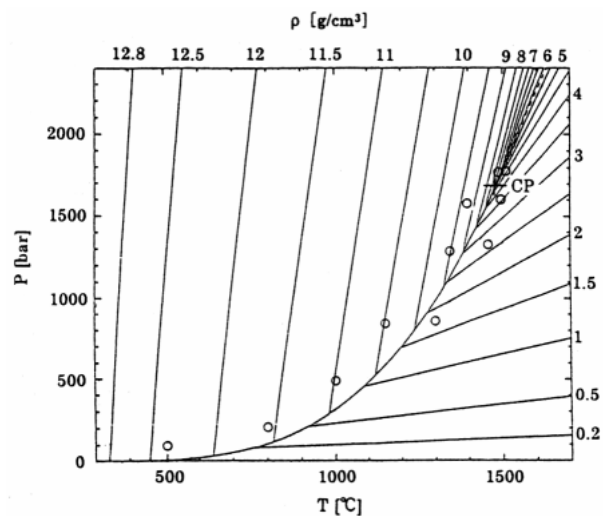

Fig.1 The density isochores of fluid Hg plotted in the pressure-temperature plane [1]. Solid line indicates the saturated vapour-pressure curve and the cross shows the critical point. Empty circles show the pressures and temperatures at which the present x-ray diffraction measurements were performed.

In the hutch we have a high pressure and high temperature generation system and an energy dispersive x-ray diffractometer. The former system includes a compressor, a high pressure vessel, thermocontroller and a chiller. Since helium gas was used as a pressure transmitting medium most of these equipments were placed in a small rooms surrounded by the protection wall built inside the hutch. The high pressure vessel permits us to measure x-ray diffraction at temperatures and pressures up to 1650°C and 2000 kg cm^{-2}. The vessel has Be windows for the incident and scattered x-rays. The high pressure vessel was mounted on the horizontal goniometer being fixed on

XZ stage.

The incident x-rays introduced into the high pressure vessel were scattered by fluid Hg.

The intensity of the outgoing scattered x-rays through Be windows was measured with a Ge solid state detector. The fluid sample was contained in the specially designed sapphire cell being transparent to x-rays and resistant to chemical corrosion by the hot fluid Hg [2]. The sample thickness was 30 m and measurements were made by the transmitting mode. X-ray diffraction measurements for fluid Hg were carried out in the temperature and pressure ranges up to 1520°C and 1775 bar along the saturated vapour-pressure curve and with densities ranging from 13.55 to 1.8 gcm^{-3}. Empty circles in Fig.1 show the temperatures and pressures at which the present measurements were made.

3.Results and discussion

Figure 2 shows the interference function, $S(k)$, of fluid Hg at different temperatures and pressures. The $S(k)$ curve at 27°C and 93 bar is in good agreement with previous data obtained by the usual angle-dispersive method. The characteristic feature of the temperature and pressure variations of $S(k)$ is that the oscillation damps and the peak width becomes broad with increasing temperature and pressure, or with decreasing density. This tendency is distinct in the first peak and completely damped in the dense vopour region. It is noticed that the data in the vapour region could be obtained. The values of $S(k)$ at low k increase with increasing temperature and pressure and remarkably in the vaour region. We have obtained the pair distribution functions, $g(r)$, by Fourier-transforming $S(k)$. The density variations of the coordination number, N_1, and the nearest neighbour distance, r_1 obtained from $g(r)$ are in good agreement with previous data [2]: The volume expansion of liquid He is not accompanied by a uniform increase of r_1 but mainly caused by a decrease of N_1, When the M-NM transition region is crossed the rate of decrease N_1, becomes small and r_1 starts to elongate. The new information obtained is as follows. The asymmetric shape of the first peak of $g(r)$ becomes much clearer, which will give more information about the first coordination. In the vapour region the first maxima of $g(r)$ are located

around 3.3-3.4 A which is close to the interatomic distance of Hg dimers. There still appear small ripples around first peak in $g(r)$ due to the termination, so the detailed data analysis is now in progress.
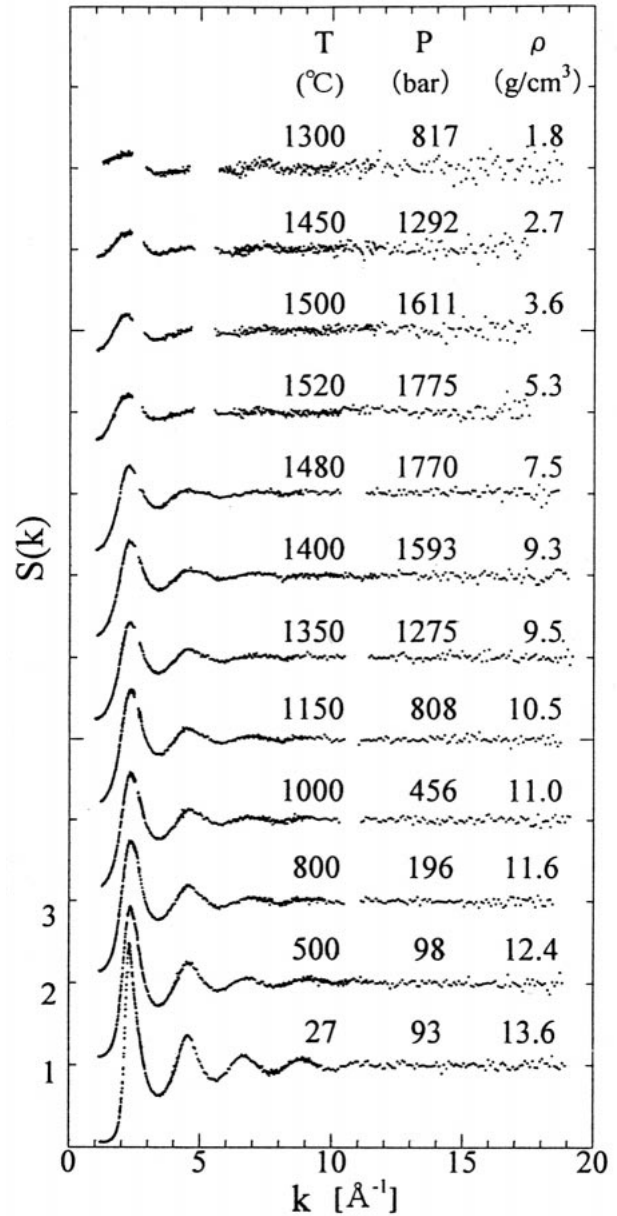

Fig.2 Interference functions, $S(k)$, for expanded fluid Hg in the temperature and pressure ranges up to 1520°C and 1775 bar along the saturated vapour-pressure curve.

References

[1] W. Gotzlaff, PhD Thesis, University of Mar-burg, 1998.
[2] K.Tamura and S.Hosokawa, J. Non-Cryst. Solids, 150 (1992) 29-34.