Electrostatic levitation furnace for x-ray diffraction measurements of high-temperature liquid materials

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Abstract: An electrostatic levitator was developed for the structural analysis of high-temperature and undercooled liquids by x-ray diffraction. The apparatus can be used to investigate the structure of metallic, semiconductor, and ceramic liquids. Samples of zirconium, silicon, and alumina in their liquid phases could be kept levitated for more than one hour with this apparatus. This was sufficiently long to perform a detailed analysis of the liquid structure by x-ray diffraction techniques.

Key words: Electrostatic levitator, X-ray diffraction, liquid structure, undercooled liquid

1. Introduction

Knowledge of the microscopic nature of matter is of paramount importance in materials science. In particular, information about the atomic configuration is essential for understanding the characteristic properties of disordered matter. Therefore, a huge amount of efforts has been devoted to the development of experimental techniques to study the structure of liquids with x-ray or neutron diffraction techniques^{1, 2, 3}. In this decade, intense and high-energy x-ray beam sources, especially synchrotron radiation, have emerged, and can be used for diffraction experiments of disordered matter. Compared to former experimental facilities, they make it possible to perform highly precise investigations of the structure of liquids in a much shorter time.

Although the methods and facilities for diffraction experiments have improved rapidly, the sample handling techniques of high–temperature liquids have not been developed at the same pace because of the difficulty in the selection of crucible materials. In the case of liquid metals, several ceramics (e.g., fused silica, sintered alumina, sapphire, graphite, boron nitride) have been used for crucibles. Despite this, the maximum temperature of the experiments has been limited by the corrosion of the crucible.

Levitation techniques use a variety of external forces (e.g., aerodynamic⁴, acoustic⁵, electromagnetic⁶, electrostatic⁷) to hold fixed in space a small amount of material without a crucible. When a levitated sample is in its liquid phase, it takes a spheroidal shape because the lack of crucible minimizes the energy of its free surface. In particular, a great deal of attention has been given to the measurement of the thermophysical properties of extremely high–temperature melts^{8,9} and the study of

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solidification phenomena from deeply undercooled liquids10.

The levitation techniques can also be a very elegant way to handle liquid samples in diffraction experiments. In that respect, they offer many advantages. The liquid sample being containerlessly processed, there is no need to subtract the diffraction contribution from a crucible. Hence, this reduces by half the measurement time because it is not necessary to measure the diffraction from the empty cell. In addition, the symmetrical shape of the sample (nearly spherical) makes it easier to evaluate the correction of absorption and multiple scattering. Moreover, and the most important advantage, the liquid samples under containerless conditions can easily reach deep undercooled states because the heterogeneous nucleation sites, which usually occur at the contact points of the crucible, are suppressed. It makes possible the observation of the structure of undercooled liquids.

Several levitation techniques have been applied to the diffraction experiments for the structural analysis of liquid matter^{11, 12, 13, 14}. The aerodynamic levitation is simple, yet, useful for such experiments. In this method, a sample is levitated in a conical nozzle at the location of the minimum potential well of the gas flow. Since the sample is small (1–3 mm dia.), this levitator is well suited for use with a high–energy synchrotron radiation x–ray beam. Moreover, it can process several types of materials under various atmospheres but can not be used under vacuum conditions. So far, x–ray diffraction experiments of several high–temperature melts (e.g., liquid boron, alumina) have been performed with this technique, allowing the determination of the static structure factors of these liquids^{11, 12}. The electromagnetic levitation is another technique applicable for diffraction experiments. In this method, a sample of conductive material is levitated in a RF coil. The high frequency current of the coil induces an Eddy's current in the metallic sample and the electromagnetic force is induced for the levitation. The levitated sample is positioned at a stabilized point which depends on the shape of the coil and on the electromagnetic properties of the sample. Since the sample size is large (6–8 mm dia.), it is especially well suited for neutron scattering experiments. Schenk et al.¹³ applied this levitation method to neutron and x–ray scattering experiments of equilibrium and non–equilibrium liquid metals.

It is also possible to levitate matter by applying electrostatic forces, through an active feedback system, on samples charged by electronic emission⁷. Electrostatic levitation is extremely attractive for x-ray diffraction experiments for several reasons. Taking into account the x-ray absorption coefficient and the atomic scattering factor of typical high-temperature metallic melts, the size of the levitated sample (1-2 mm dia.) is suitable for the diffraction of high-energy x-rays from synchrotron radiation source. In addition, the charged liquid sample being levitated between pairs of electrodes, the sample is free from any obstacle, such as the nozzle or the coils in other levitators. Moreover, to avoid electrical breakdown when applying a high voltage between electrodes, electrostatic levitators have to be operated either under pressurized atmospheres (~ 4 KPa) or under high vacuum. The high-vacuum conditions are particularly appealing for x-ray diffraction because there is no need to consider the scattering from the ambient gas. Recently, Gangopadhyay et al. 4 used such levitators for x-ray diffraction with a synchrotron radiation facility and observed the static structure factors and solidification behavior of several metallic melts. Electrostatic levitation has also been applied by Aoki et al.15 to neutron diffraction experiments and successfully used to measure the diffraction pattern of sintered alumina at room temperature. Although the validity of electrostatic levitation for diffraction experiments has been recognized, the previous facilities exhibited limitations for precise measurements. In particular, the observable range of the diffraction angle is limited which affects the resolution of the data obtained through a Fourier transform. The atomic configuration of liquids in real space can be investigated from the radial distribution function, g(r), which is obtained by a Fourier transformation of S(Q) as follows;

$$g(r) = 1 + \frac{1}{2\pi^2 or} \int_0^\infty \left[S(Q) - 1 \right] Q \sin Qr \, dr, \tag{1}$$

where ρ is the number density, S(Q) is the static structure factor, and Q is the momentum transfer. The g(r) obtained from

diffraction experiments bears experimental errors essentially because the Q range of S(Q) is limited due to the wavelength of x-rays, the diverted tail of the direct beam, and the observable range of 2θ . In addition, the previously developed electrostatic levitator could be used only on large beam source facilities (e.g., synchrotron radiation facilities or nuclear reactors 15). Therefore, this restricts the opportunities for experiments because of the limited machine time.

In this study, we developed an electrostatic levitator for x-ray diffraction measurements with a high flexibility of beam sources. Since the system is very compact, it can be utilized not only with the diffractometer at the high-energy x-ray diffraction beamline, BL04B2 of the synchrotron radiation facility, SPring-8%, but also with a laboratory x-ray system (RI-GAKU RINT). The electrostatic levitator was designed for the x-ray diffraction measurements with a two-axis diffractometer with slits collimation coupled with a germanium detector or a proportional counter. The scattering intensity of each scattering angle, 2θ , was acquired by the counter with the step-scan method. The high-energy x-ray beam from the synchrotron source being higher than 100 keV, is a very attractive probe for the liquid structure analysis compared to laboratory x-ray sources. The structure factor of the bulk liquid can be measured using the high-energy x-rays due to the high penetration of incident x-ray even for samples of 3d or 4d transition metals. In addition, the momentum transfer, $Q = 4p \sin\theta/\lambda$ (2θ : scattering angle, λ : wavelength of incident x-rays), being proportional to the x-ray energy, the static structure factor, S(Q), in sufficiently wide Q range, can be obtained from the measurement of diffraction pattern with small scattering angles. On the other hand, the laboratory x-ray source can be used for diffraction experiments of lighter materials, such as silicon. Since the laboratory x-ray source is free from the restriction of user time of the facility, preliminary or challenging experiments can be performed with trial and error, which complements synchrotron x-ray experiments.

This report describes the development of the system and presents the results of a preliminary application for the atomic structure analysis by x-ray diffraction measurements.

2. Electrostatic Levitation for the Structural Analysis by X-ray Diffraction Technique

The design of the present apparatus was based on an electrostatic levitator which was developed by Rhim et al. but optimized for the liquid structure analysis of high-temperature melts by x-ray diffraction technique. The apparatus consisted of a vacuum chamber, a sample position control system, and a sample heating source. The sample, charged by electronic emission, was levitated by applying an electrostatic field (typically 20 to 30 kV/cm for metallic materials) between two electrodes. To prevent the electrical breakdown, the electrodes were contained in a chamber that was evacuated to a level of vacuum better than 1 x 10⁻⁴ Pa with a turbo molecular pump attached directly to the side of the chamber. Figures 1 and 2 illustrate the side and top views of the chamber, respectively. The chamber has a cylindrical shape (height: 200 mm; diameter: 200 mm) and comprises several view ports. A thin sapphire window (thickness: 0.5 mm; diameter: 17 mm) allowed the incident x-ray beam to reach the sample and a rectangular and curved beryllium window permitted the detection of the intensity of the x-rays diffracted by the levitated sample over a wide angle. The available range of 2θ was -5 to 80 degrees, which is wider than that previously reported¹³. Sufficiently wide Q range $(Q \sim 11.5 \text{ Å}^{-1})$ can be obtained even for laboratory x-ray source (Mo Ka). Five silica glass windows, located on the top of the chamber, were used, along with mirrors inside the chamber, for the position control system and sample observation by a camera. A ZnSe window (or lens) in the middle of the top plate was used for the sample heating by a CO₂ laser (wavelength: 10.6 μm; max. power: 240 W). A glass window on the side of the chamber was employed for the temperature measurement by a single-color pyrometer. Two valves located on the top plate acted as an air lock that enabled to insert samples without breaking the vacuum.

The design of the electrodes is of the utmost importance for electrostatic levitators. In our levitator, there were two main electrodes for vertical and horizontal control and four side electrodes for additional horizontal control. The main electrodes consisted in two parallel disks. The upper electrode (40 mm dia.) was suspended from the top plate using insulating ceramic

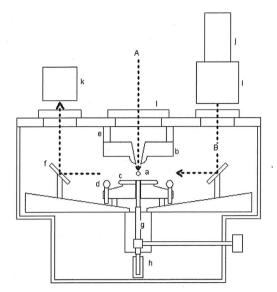
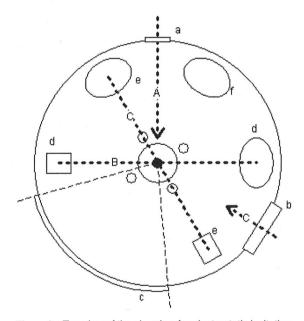


Figure. 1 Side view of the chamber for electrostatic levitation.

a: levitated sample, b: upper electrode, c: lower electrode, d: side electrodes, e: ceramic support, f: mirrors, g: positioning rod, h: solenoid, A: beam path of heating CO₂ laser, B: beam path of positioning He–Ne laser.



 ${\bf Figure~2}~~{\bf Top~view~of~the~chamber~for~electrostatic~levitation}.$

a: sapphire window for the incident x-ray beam, b: glass window for the pyrometer, c: beryllium window for diffracted x-rays, d: mirrors for the He-Ne laser for positioning in X-Z directions, e: mirrors for the He-Ne laser for positioning in Y direction, f: mirror for CCD camera, A: path of incident x-rays, B&C path of He-Ne lasers, C: path of pyrometer.

rods, that was connected to a high voltage amplifier. It had a spherical end which generated a concave electrostatic field that helped to stabilize the sample laterally. In addition, a through hole (3 mm dia.) in its center allowed sample heating by the CO₂ laser. The lower electrode (20 mm dia.) was electrically grounded and had a hole allowing sample handling by a positioning rod. This rod can be moved up and down from the outside of the chamber to set the initial position of sample. It was equipped with a small tubular solenoid that was used to ease sample initiation levitation (cf. Section 3). Four small spherical

electrodes were distributed around the lower electrode for additional control of the sample position along the horizontal direction.

To maintain a stable sample levitation, the voltage of the electrodes was controlled actively with a position sensing system, a computer, and high-voltage DC amplifiers. The position of the levitated sample was detected by two sets of position sensors and associated He-Ne lasers. In each set, the expanded He-Ne laser beam (10 mm dia.) illuminated the sample and its shadow was projected on the position detector which was located on the opposite side of the He-Ne laser. The computer received an electric signal from the position detectors that corresponded to the sample position. It then calculated the control signal using a PID control scheme, and sent the proper information to the voltage amplifier that changed the voltage of the electrodes. By doing this at a feedback rate of 1000 Hz, the sample could maintain a fixed position. The position control system used in this study was similar to that reported elsewhere⁷ but the optical paths for position sensing were modified because of the constraint of x-ray scattering. The He-Ne lasers and the position detectors were located on the top plate and therefore, the optical paths of the lasers were bent twice by mirrors. This optical configuration offered a wide observation view of the sample as well as helped to miniaturize the chamber, making it easier to set up at the synchrotron radiation facility.

3. Experimental

Present research was performed to verify the applicability of electrostatic levitation methods to the x-ray diffraction measurements. Electrostatic levitation is, in principle, applicable to a wide variety of materials because all charged materials can be levitated by the action of electrostatic forces. For the first experiment, zirconium was selected and the structural analysis of its liquid phase was carried out by high-energy x-ray diffraction measurement at SPring-8, which is the 3rd generation synchrotron radiation facility in Japan. Similar experiments were performed with molten silicon and alumina samples using a laboratory x-ray source.

For the present experiments, the typical sample size was about 2 mm in diameter. Spheroid zirconium samples were made by cutting 99.5 Wt. % pure zirconium wire into 30–32 mg pieces. The pieces were melted with a diode laser (wavelength: 808 nm; max. power: 200 W) in a glove box filled with purified argon. The melted pieces adopted a spheroidal shape spontaneously because of the surface tension. Silicon and alumina samples were made similarly.

Heating is the most delicate task when processing a sample with electrostatic levitators since the sample charge has a tendency to decrease due to the evaporation of absorbed gas or metallic oxides from its surface. In particular, heating of the levitated sample from room temperature must be done carefully since the sample charge starts to escape at a temperature of about 800 K. However, the charge can be increased through electronic emission if the sample reaches a temperature at which thermionic emission dominates (~ 1500 K in the case of metals). In order to overcome these difficulties, the "hot launch" method was used¹⁷. To initiate levitation, the sample was heated to remove surface oxides. When it reached ~ 1500 K, thermionic emission dominated and the high voltage was applied to the electrodes and the feedback control system was activated. Once levitated, the sample could then be brought to temperatures beyond the melting point or be maintained under undercooled conditions for hours. However, low melting point materials (e.g., Si) have a tendency to stick to the positioning rod and, therefore, the heated sample must be tossed while heating. In the present apparatus, a small solenoid which created vibrations, was fixed at the lower part of the rod. The solenoid was activated remotely during the monitoring of the sample temperature.

The temperature of the sample was measured with the use of a single-color pyrometer. The emissivity of the sample is necessary to obtain the exact value of the temperature. However, the emissivity strongly depends on the sample status, the sample size, the focus of collimation lens of the pyrometer, and the transparency of the window of the chamber. In the pre-

sent research, the emissivity was calibrated with the melting points of zirconium and silicon. The undercooled liquid state was established simply by decreasing the laser power. After a certain depth of undercooling was reached, the sudden increase of the sample temperature due to the release of the latent heat of fusion was observed at solidification. Therefore, the undercooled liquid state was confirmed by monitoring the signal of the pyrometer. The laser power was controlled to keep constant the temperature of the sample.

The high-accuracy measurement of the liquid structure is one of the major purposes of this research. The two-axis diffractometer is the most typical instrument used nowadays for the diffraction measurements. The x-ray source was selected considering the absorption coefficient of the material. For Zr, which has a high absorption coefficient, the high-energy xrays (113 keV) from the BL04B2 of SPring-8 were used. For silicon and alumina, the laboratory x-rays from a Mo rotation target were sufficient to carry out preliminary diffraction experiments. The size of the incident beam was 0.7 mm in width and 2.0 mm in height for the synchrotron radiation x-rays, and 3 mm x 3 mm for the laboratory x-rays. The incident beam was collimated by the slit and delivered to the vacuum chamber through a sapphire window. The angular dependence of the intensity of the x-rays diffracted from the sample was measured in transmission geometry by a Ge detector or a proportional counter with a graphite monochrometor. Slit collimation eliminated the scattering from the windows on the chamber, which allowed precise measurements of the diffraction from the sample. The intensity of diffracted x-rays was acquired in each diffraction angle by the step scan method. The diffraction data was collected over a 2θ range of 0.3-25 degrees for measurements with synchrotron radiation and 0.5-80 degrees for measurements with laboratory x-rays. The obtained Q range of S (O) was 0.3 - 24.7 Å⁻¹ for the synchrotron radiation and 0.08 - 11.4 Å⁻¹ for the laboratory X-ray. The duration of acquisition of each diffraction angle was greater than 5 seconds which is sufficiently long for high statistics. In order to obtain the static structure factor of liquids, data correction of the absorption, background, polarization, and multiple scattering must be taken into account^{1,2,3}. The width of incident x-rays (0.7 mm) being narrower than the sample diameter (2 mm) in the synchrotron radiation experiments, however the influence of total angular dependence of the absorption coefficient was negligibly small because diffraction experiments can be performed with rather small scattering angles with high-energy x-rays. In addition, it is worth mentioning that the absorption coefficient itself is very small with high-energy x-rays (mass absorption coefficient is 0.673 for zirconium21), which implies that the contribution of absorption correction is extremely small. The contribution of multiple scattering has been mentioned for the structural analysis of disordered matter not only for the neutron scattering experiments but also for x-ray diffraction measurements1. In the present study, the sample was spherical and quite small. We evaluated the contribution of double scattering compared to that of the single scattering following the method reported by Warren¹. The ratio of the double scattering to the single scattering in the present case was less than one percent, and therefore, the contribution was neglected.

After correcting for absorption²¹, background, and multiple scattering, the contribution of Compton scattering²² was subtracted and then the x-ray-weighted static structure factor²³, S(Q), was derived from the corrected coherent intensity, I(Q), according to

$$I(Q) = \left| \left\langle f(Q) \right\rangle \right|^2 \left[S(Q) - 1 \right] + \left\langle \left| f(Q) \right|^2 \right\rangle, \tag{2}$$

where the angular brackets represent averages over all atoms and f(Q) is the atomic form factor²⁴.

4. RESULTS AND DISCUSSION

The zirconium, silicon, and alumina samples were representative materials for metals, semiconductors, and ceramics, respectively. All of the samples could be levitated successfully in their molten states and could be maintained at a fixed temperature for more than one hour, which was sufficient for the measurement of x-ray diffraction. The fluctuation of the samples could be representative materials for metals, semiconductors, and ceramics, respectively.

ple position was less than 0.1 mm for all materials during measurements. The diffracted x-rays were counted for five seconds for each diffraction angle. In addition, in the case of the laboratory x-ray facility, the area of the incident x-ray beam (3 mm x 3 mm) was much larger than the cross sectional area of the sample. Therefore, the sample fluctuation did not affect the diffraction data. The diffraction patterns for all three materials are shown in figures 3, 4 and 5. As can be seen in figure 3 and 4, diffraction from the empty chamber was almost negligible at Q > 1 Å⁻¹, since the use of vacuum and proper shields to the detector effectively suppressed the background. Therefore, the error from the background correction was extremely small, which is remarkably different from conical nozzle levitation. Thus the combination of containerless conditions and high vacuum enabled us to obtain very reliable observation of the liquid structure.

The static structure factor S(Q) of liquid samples can be obtained from the diffraction intensity. The static structure factors shown in figures 6 to 8 demonstrated that we have succeeded in performing precise observations of the liquid structure with the present apparatus, using not only using synchrotron radiation x-rays but also laboratory x-ray source. The liquid structures of the materials investigated in this study have been measured with other types of levitators in conjunction with

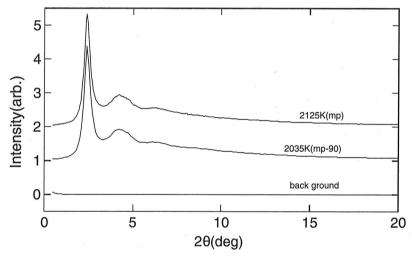


Figure 3 High-energy x-ray diffraction pattern of liquid and undercooled zirconium.

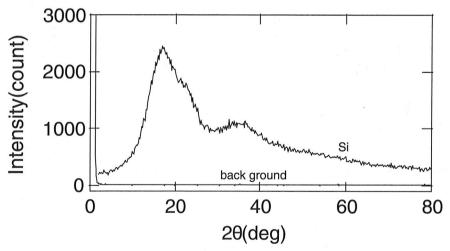


Figure 4 X-ray diffraction pattern of liquid silicon at the melting point.

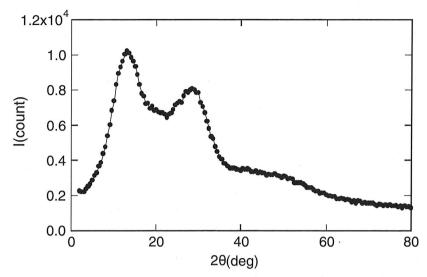


Figure 5 X-ray diffraction pattern of liquid alumina at the melting point.

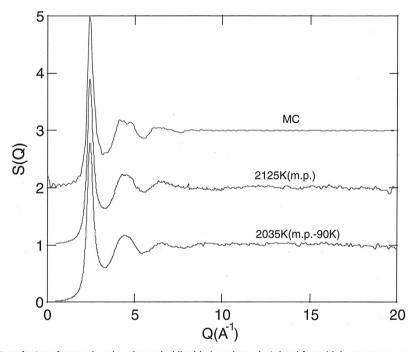


Figure 6 Static structure factor of normal and undercooled liquid zirconium obateined from high–energy x–ray diffraction experiment and Monte Carlo simulation.

neutron or x-ray scattering. Our experimental results are in good agreement with those previously published^{13, 18, 19, 20}. However, the quality of data, such as the Q range of S(Q) and low background, was significantly improved. The S(Q) of liquid zirconium was observed in the range of Q=0.5 to 20.0 Å, which was wider than that of previous research¹³.

To demonstrate the quality of the obtained S(Q), the effective pair potential, $u_{eff}(r)$, was deduced based on the Modified Hypernetted Chain approximation²⁵, as follows:

$$u_{eff}(r)/k_B T = g(r) - 1 - c(r) - \ln g(r) + B_{HS}(r, \eta), \tag{3}$$

where c(r) is the direct correlation function, k_B is the Boltzmann constant, and T is the temperature. The c(r) was calculated

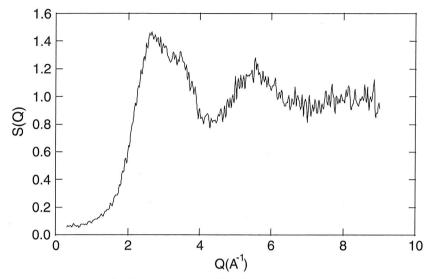


Figure 7 Static structure factor of liquid silicon at the melting point.

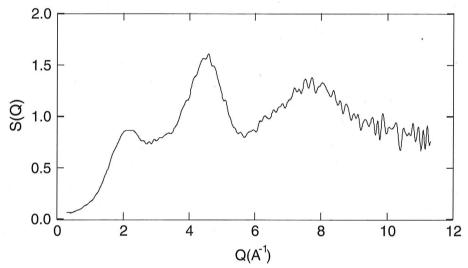


Figure 8 Static structure factor of liquid alumina at the melting point.

from S(Q), as follows:

$$c(r) = \frac{1}{2\pi\rho r} \int \left(1 - \frac{1}{S(Q)}\right) Q \exp(-iQ \cdot r) dQ. \tag{4}$$

The reliability of S(Q) in the small Q region is quite important for this calculation because the Fourier transform of the term of 1/S(Q) must be calculated. In addition, high–Q data are necessary to reduce the truncation error in the Fourier transform of S(Q) to g(r) and c(r), which is necessary for the precise determination of the repulsive part of $u_{eff}(r)$. The $B_{HS}(r, \eta)$ is the bridge function of the hard sphere fluid and η is the packing fraction. For the conventional estimation of B_{HS} , the η of liquid zirconium was taken as 0.46, which is generally used for the packing fraction at the melting point for hard sphere fluids²⁶. The $u_{eff}(r)$ obtained was shown in figure 9. For the verification of $u_{eff}(r)$, the liquid structure was reproduced by using a Monte Carlo simulation^{27, 28} (MC) with the $u_{eff}(r)$. The temperature of MC was 2125 K. The S(Q) derived from MC is in good agreement with the experimental results as can be seen in figure 6. The $u_{eff}(r)$ is widely applicable for the evaluation of not only the

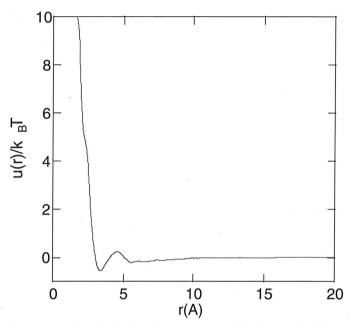


Figure 9 Effective pair potential of liquid zirconium based on modified hypernetted chain approximation.

static properties but also the dynamic properties. For example, transport properties, such as self-diffusion and viscosity coefficients, can be estimated from the combination of $u_{eff}(r)$ and molecular dynamics simulation (MD). The viscosity coefficient of liquid zirconium has been measured by the oscillation drop method coupled with the electrostatic levitator²⁹. The detail analysis of viscosity coefficients obtained from MD with $u_{eff}(r)$ is in progress.

In the case of laboratory experiments, the background can be removed from the measurements and reasonable S(Q) can be obtained with the use of only standard procedures of data corrections. The S(Q) of liquid silicon measured with laboratory x-rays, the Q range of S(Q) was rather small for the calculation of g(r) by the simple Fourier transform as given by eqn (1). Nevertheless, the present S(Q) for liquid silicon was sufficiently good to be applicable for the structure analysis. For example, the g(r) of liquid silicon was calculated by using the Reverse Monte Carlo³⁰ (RMC) simulation and the coordination number of nearest neighbors was estimated. The coordination number was 5.9 which agrees well with previous research¹⁹. In addition, the present S(Q) of liquid alumina shows a better agreement with the molecular dynamics simulation²⁰ than with the experimental data reported by Krishnan et al²⁰. This can be explained by the fact that the reliability of our S(Q) in the low Q region is much better than the experimental data reported in ref. 20. The structural difference in liquid alumina depending on the ambient atmospheric oxygen concentration was reported previously²⁰ but no out such behavior was found in our data. Furthermore, our preliminary high-energy x-ray diffraction experiments using a conical nozzle did not exhibit such a behavior, suggesting that the structural difference of liquid alumina in oxidizing and reducing condition is still an open question. Diffraction experiments on liquid silicon and alumina with synchrotron radiation x-rays are planned in a near future.

Different types of materials could be successfully processed using a single apparatus. For the diffraction experiments, this feature is a great advantage because a common optical set—up and background calibrations can be used. We developed an electrostatic levitator which is applicable to a wide variety of materials and x—ray sources. Furthermore we confirmed that the quality of data of high—temperature liquids is much better than those of previously published data. We believe that we overcame the problem of quality of data of high—temperature and undercooled liquids. The present apparatus will be able to perform the experimental analysis of high—temperature melts with high precision and will contribute to the fundamental un-

derstanding of the nature of liquids in normal and undercooled states.

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