Int J Thermophys (2008) 29:2006–2014
DOI 10.1007/s10765-008-0533-7

Precise Density Measurements for Electromagnetically Levitated Liquid Combined with Surface Oscillation Analysis

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Published online: 18 November 2008

Abstract A new method is proposed for accurately measuring the densities of high-temperature liquids which involves analyzing the surface oscillations of levitated droplets. This method makes it easy to improve on the accuracy of density measurements obtained by using conventional electromagnetic levitation systems. In addition, the errors in density measurements made on the ground are further reduced by applying a static magnetic field to suppress surface oscillations in levitated liquid droplets. The magnetic field interacts with electrical currents in the levitated droplet, thereby generating a Lorentz force; this force suppresses flow within the liquid droplet. By combining both these methods, the scatter in density measurements for molten Si at temperatures in the range from 1,500 K to 1,900 K is reduced by an order of magnitude compared with previously reported data. Using this new method, the density of molten SiGe has been measured at temperatures from 1,350 K to 1,650 K.

Keywords Density · Electromagnetic levitation · Molten Si · Molten SiGe · Surface oscillations

1 Introduction

Electromagnetic levitation (EML) is a useful method for measuring the thermophysical properties (e.g., density, surface tension, and viscosity) of molten metals and

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semiconductors at high temperatures [1]. The EML technique has large advantages for keeping liquids in a supercooled state and for maintaining the high-temperature state while maintaining the high purity of the measurement samples owing to the containerless environment. This makes it possible to precisely determine the temperature dependence of the thermophysical properties of molten samples over a wide temperature range. Due to these advantages, EML is expected to be used in space environments for accurately measuring the thermophysical properties of molten materials [2].

When using EML to measure the density of molten metals or semiconductors, the volume of the levitated droplet must be precisely determined from its shape. In conventional methods, the volume of the droplet is obtained by averaging measurements from several hundred images; when using these methods, however, large errors in temperature-dependent data are caused due to the asymmetry of the surface oscillations of electromagnetically levitated liquid droplets [3]. Especially for measurements performed on the ground, the large electromagnetic force required to levitate a sample enhances surface oscillations, resulting in large errors in volume measurements. To reduce these errors, the asymmetry of surface oscillations must be suppressed.

One method to achieve this is to apply a magnetic field [4]. The magnetic field interacts with electrical currents in a levitated sample, thereby generating a Lorentz force; this force suppresses flow within the liquid droplet. However, a specially designed magnet and/or EML system must be used to suppress asymmetrical surface oscillations of samples levitated by EML. Therefore, a new method is proposed for accurately measuring the densities of high-temperature liquids, which involves analyzing the surface oscillations of droplets. This method makes it easy to improve on the accuracy of density measurements obtained by using a conventional EML system. To verify this, we investigated whether combining surface vibration analysis with the application of a static magnetic field to suppress surface oscillations reduced the scatter of density measurements of molten Si. Using these techniques, we measured the density of molten $Si_{50}Ge_{50}$, which has been measured previously only at 1,715 K [5]. Accurate measurements of its density will be vital for making materials from $Si_{50}Ge_{50}$. This is important because $Si_{50}Ge_{50}$ is expected to be used in the next generation of solar cell devices.

2 Experiments

Electromagnetic levitation experiments were performed using an rf power supply at 250 kHz and 10 kW using the arrangement shown in Fig. 1. Si and $Si_{50}Ge_{50}$ solid samples were set on a carbon rod coated with a pyrolytic boron nitride (PBN) cap in a vacuum chamber. The chamber was evacuated to 10^{-4} Pa and then backfilled with Ar gas with 6N purity. The profile of the levitated sample was observed from the side using a high-speed camera (MotionScope, PCI 1000S) with a frame rate of 250 Hz. Each image of the levitated sample was recorded with a resolution of 256×256 pixels. To calibrate the length from digital images, we recorded an image of a 5 mm diameter sphere of solid silicon. The diameter of the silicon sphere is accurate to 0.01 mm. Two polarization filters were used to ensure that the brightness of the sample remained approximately constant with temperature. The temperature of

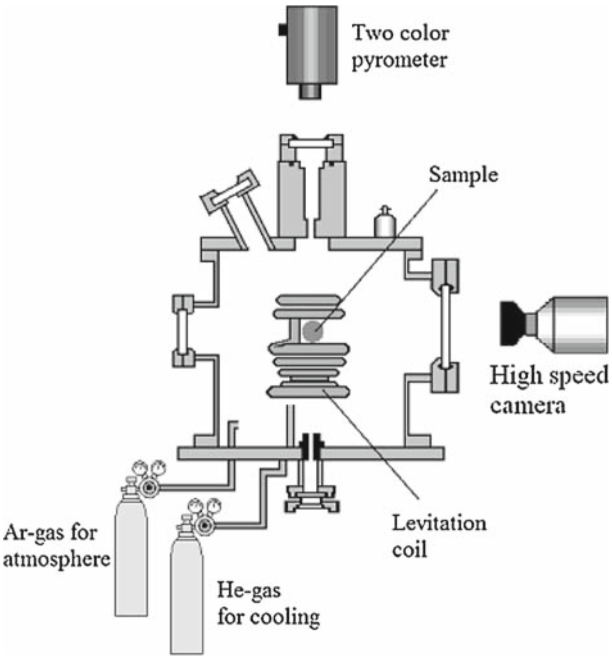

Fig. 1 Schematic diagram of electromagnetic levitation system for measuring the density of molten samples by image processing of the images obtained using a high-speed camera

the levitated sample was measured with a two-color pyrometer (CHINO, IR-CAQ) at wavelengths of 850 nm and 1,000 nm using a value of 1.0 for the emissivity ratio [6].

The density measurements were performed using an image processing method [3], in which the volume of the sample is determined from its profile. The mass of the sample was measured before and after levitation in order to account for any volume change due to evaporation of the sample. Finally, the density was calculated by dividing the mass by the volume. Using this method, we measured the densities of molten Si and $Si_{50}Ge_{50}$. To confirm the effectiveness of surface oscillation analysis on density measurements of levitated liquid samples, we also applied a static magnetic field and measured the volume of levitated samples to determine how much the magnetic field suppressed the surface oscillations. This suppression of the surface oscillations an electromagnetically levitated droplet is caused by the Lorentz force that is generated by the static magnetic field, and it facilities the measurement of the density because the shape of the droplet is approximately that of a solid body. We used a superconducting magnet, and the EML system was constructed inside the magnet. Details of the EML system in the superconducting magnet are described elsewhere [7–9].

3 Results and Discussion

The volume of the levitated samples was obtained by the following procedure. Figure 2a shows a photograph of the levitated molten Si taken by a high-speed camera.

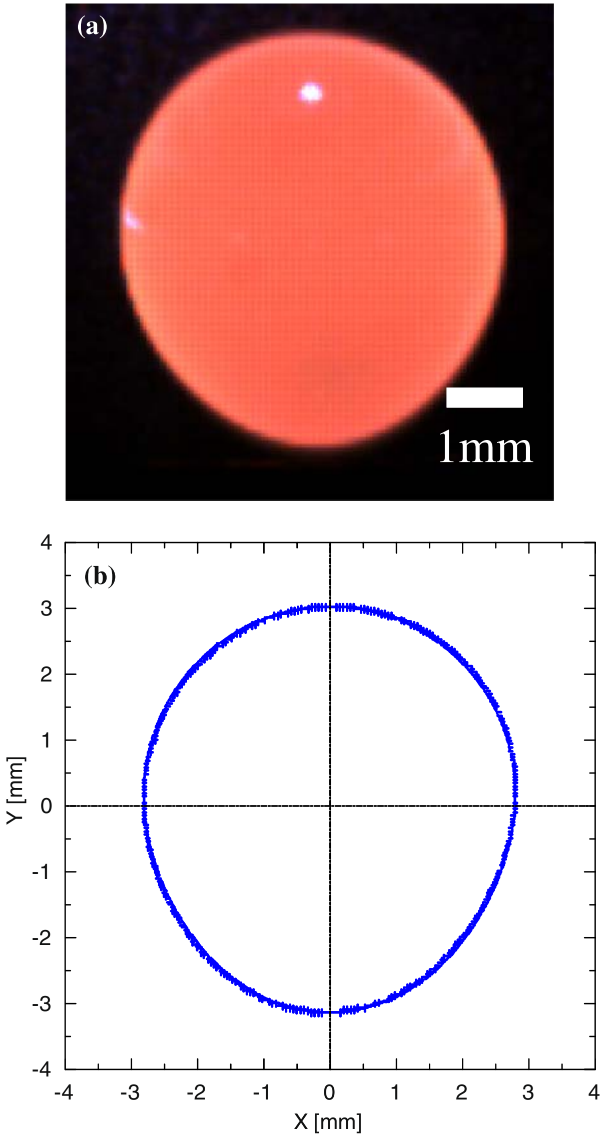

Fig. 2 (**a**) Photograph of electromagnetically levitated molten Si taken by a high-speed camera and (**b**) edge picture of levitated molten Si obtained from the photograph shown in (**a**) by image processing

An image processing procedure was used to detect the edge of the droplet in each image. Figure 2b shows an image of the edge of the levitated molten Si droplet shown in Fig. 2a. The length r from the center of gravity of the droplet to the edge was then fitted by a series of Legendre polynomials $P_n(\cos(\theta))$ of order six, as given by

$$r\,(\cos(\theta)) = \sum_{n=0}^{6} a_n P_n(\cos(\theta)). \tag{1}$$

The droplet at static equilibrium is assumed to be rotationally symmetric about the vertical axis. The volume V of the droplet was calculated using the equation,

$$V = \frac{2\pi}{3}\int_{-1}^{1} r^3(\cos(\theta))\mathrm{d}\cos(\theta). \tag{2}$$

However, the shape of the droplets, levitated by an electromagnetic force, fluctuated due to the surface oscillations enhanced by the electromagnetic force. The three-dimensional shape of an oscillating droplet is given by

$$r = R + \varepsilon Y_l^m(\theta, \varphi) = R + \varepsilon P_l^m(\theta)\cos(m\varphi)\cos(\omega_l t) \tag{3}$$

where R is the radius of a sphere, ε is the oscillation amplitude, ω is the frequency of surface oscillations, and l and m are the labels that indicate the mode of the oscillation. Thus, the apparent volume of the levitated droplets observed from the side periodically changed with time, as shown in Fig. 3, which gives the results for molten Si at 1,680 K. We analyzed the periodic change in the apparent volume of the droplets with surface oscillation analysis used for surface-tension measurements [2,10]. The oscillation frequency (ω_l) of a liquid droplet is calculated by Rayleigh's equation [11],

$$\omega_l^2 = l(l-1)(l+2)\frac{4\pi}{3}\frac{\gamma}{M} \tag{4}$$

where γ is the surface tension and M is the mass of the droplet. This frequency was calculated by assuming that the droplet is spherical. However, in the case of electromagnetically levitated droplets, the shape is not completely spherical, so that the surface oscillation frequency is split into three frequencies $\omega_{2,0}$, $\omega_{2,\pm 1}$, and $\omega_{2,\pm 2}$ as given by Cummings and Blackburn [12]. The surface oscillation frequency is used

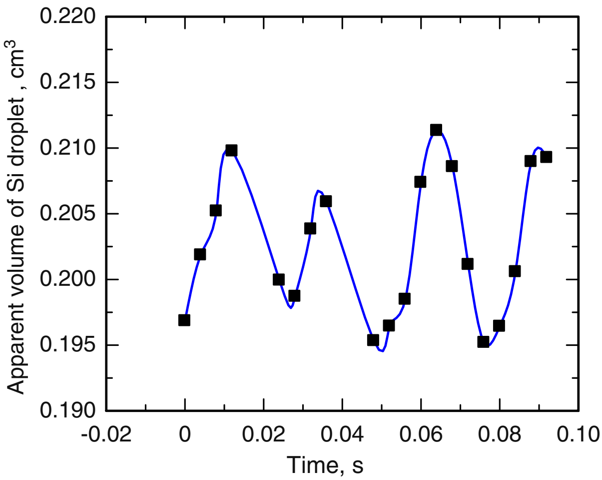

Fig. 3 Time dependence of apparent volume of electromagnetically levitated molten Si at 1,680 K

for surface-tension measurements of a levitated droplet. Moreover, the deviation of the droplet shape by the surface oscillations leads to a change in the apparent volume of the droplet. We found from this analysis that the periodic change in the apparent volume is caused by the $(l, m) = (2, 2)$ mode. For this mode, the shape of the droplet becomes rotationally symmetric with time. Thus, the volume of the droplet calculated from the side profile also changes with time. When the surface of the droplet oscillates, the true volume of the droplet is calculated from the profile in which the difference between the major and minor axes of its elliptical shape is a minimum. The true volume is obtained from the image obtained halfway between the time of the local maximum and the time of the local minimum. In the case of a levitated sample that rotates around the vertical axis, the oscillation affects only the height of the peaks in the variation of the apparent volume. In other words, if there are peaks only due to the $(l, m) = (2, 2)$ mode in the change of the apparent volume, then this demonstrates that other oscillation modes are not present on the sample surface. Therefore, we selected the profile that represents the true volume of the droplet from all the images taken by the high-speed camera.

On the basis of this analysis, we were able to obtain the true volume of a levitated droplet at a constant temperature. This analysis enabled us to reduce by one order of magnitude the scatter in the results for the density of molten Si at temperatures from 1,500 K to 1,800 K relative to previous results obtained using EML [3,13]. We confirmed that this reduction in the scatter of the density data for molten Si is associated with damping of surface oscillations by the static magnetic field. Onishi et al. [14] reported a reduction in the scatter of measured density values of molten Si in the temperature range from 1,370 K to 1,770 K due to damping of surface oscillations caused by applying a static magnetic field of 6 T. However, they did not provide information on the minimum magnetic field strength required to damp the surface oscillations of electromagnetically levitated molten Si. Therefore, in order to determine this magnetic field strength, we applied static magnetic fields from 0 T to 2 T. Figure 4 shows the apparent volume of electromagnetically levitated molten Si for various static magnetic fields. This figure shows that in a static magnetic field of 0.25 T the volume of the molten Si droplet changed periodically in a manner similar to that when no static magnetic field was applied, but in a static magnetic field of 1.5 T, the molten Si volume did not change. This means that the surface oscillations of electromagnetically levitated molten Si droplets can be reduced by applying a static magnetic field of greater than 1.5 T. When a static magnetic field was applied, we obtained the same temperature dependence for the molten Si density [9] with the same deviations as for the method using surface oscillation analysis. Therefore, we confirmed that surface oscillation analysis can be used to precisely determine the temperature dependence of the liquid density with smaller deviations than those with previously developed methods.

We used the same method to measure the density of molten $Si_{50}Ge_{50}$ as was used for molten Si. The temperature dependence of the density of molten $Si_{50}Ge_{50}$ has not been previously reported. Figure 5 shows the change in the apparent volume of molten $Si_{50}Ge_{50}$ at 1550 K. This result shows that the apparent volume change variation of molten $Si_{50}Ge_{50}$ differs from that of molten Si due to their different surface tensions and viscosities. The results for density measurements of molten $Si_{50}Ge_{50}$ at temper-

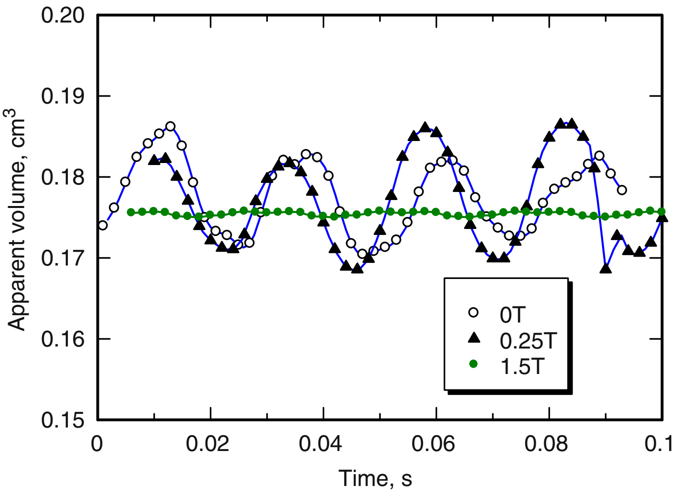

Fig. 4 Time dependence of apparent volume of electromagnetically levitated molten Si at 1,680 K under various static magnetic fields

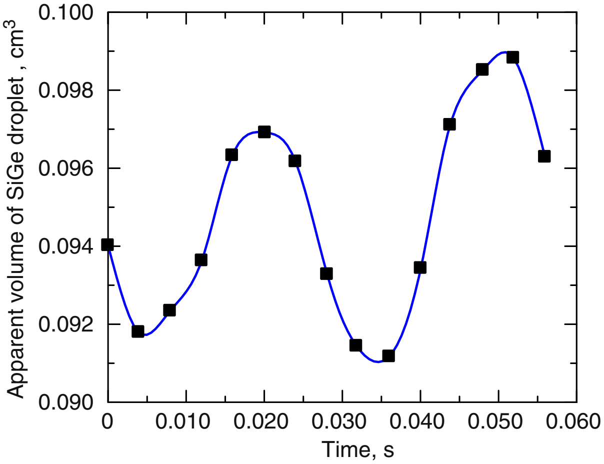

Fig. 5 Time dependence of apparent volume of electromagnetically levitated molten $Si_{50}Ge_{50}$ at 1,550 K

atures from 1,500 K to 1,800 K are shown in Fig. 6. This result shows that the scatter in the density data over this temperature range is approximately 1 %, and that the temperature dependence of the density is given by

$$\rho(T) = 4.18 \times 10^3 - 0.546 \times (T - T_m)^3 \text{ kg} \cdot \text{m}^{-3},$$

where T_m is the liquidus temperature. These represent the first reported measurements of the temperature dependence of molten $Si_{50}Ge_{50}$. $Si_{50}Ge_{50}$ is expected to be used in the next generation of solar cell devices. Thus, these data are important for making materials from $Si_{50}Ge_{50}$.

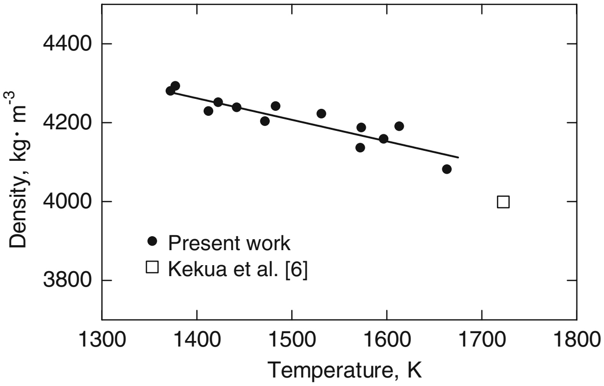

Fig. 6 Temperature dependence of the density of molten $Si_{50}Ge_{50}$. (□) Value reported by Kekua et al. [5]

4 Conclusion

We proposed a new method for measuring the precise density of molten metals and semiconductors by using an EML technique in combination with surface oscillation analysis. We confirmed a one order of magnitude reduction in the scatter of density measurements of molten Si at temperatures ranging from 1,500 K to 1,900 K. We used this new method to measure the density of molten SiGe. In the future, we intend to analyze more precisely volume fluctuations of levitated droplets including rotating the sample to make comparisons with the shape of electromagnetically levitated droplets in static magnetic fields.

Acknowledgments We would like to thank Prof. T. Hibiya of Keio University and Dr. S. Ozawa of Tokyo Metropolitan University for useful discussions about surface oscillations of liquid droplets. This study was supported by Research and Development to Promote the Creation and Utilization of an Intellectual Infrastructure from New Energy, and also by the Industrial Technology Development Organization (NEDO) with a Grant-in-Aid for Scientific Research (KAKENHI No. 19560747).

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