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ANALYSIS OF SURFACE OSCILLATION OF DROPLET UNDER MICROGRAVITY FOR THE DETERMINATION OF ITS SURFACE TENSION

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Abstract—Shape changes in a levitated droplet can affect the surface oscillations and hence the apparent surface tension of the droplet. The effect of those changes was investigated using the electromagnetic levitation method under microgravity. Microgravity conditions were obtained using the drop-shaft at the Japan Microgravity Center (JAMIC). The results established that the surface oscillation of a droplet is much simpler under the 10^{-5} G condition than under 1 G. The droplet shape was controlled by changing the current ratio between the quadrupole coil and the dipole coil. When the droplet shape is (a) spherical only one peak is observed, (b) is close to a sphere, but slightly distorted, five peaks are observed and (c) the droplet is significantly distorted, a particular oscillation mode is usually excited and only one peak is sometimes observed. However, for the third condition, the surface tension values derived from the frequency of the single peak are erroneous. *© 2000 Acta Metallurgica Inc. Published by Elsevier Science Ltd. All rights reserved.*

Keywords: Surface tension; Liquid silicon; Semiconductor; Surfaces & interfaces

1. INTRODUCTION

The levitated drop method has several advantages in measuring the surface tension of high temperature materials. In this method, the surface tension is calculated from the frequency of the surface oscillation of a levitated sample using Rayleigh's equation [1]:

$$\gamma = \frac{3}{8}\pi M \nu_R^2 \qquad (1)$$

where ν_R is the frequency of the surface oscillation, M is the droplet mass, and γ is the surface tension. The first advantage of this method is that for the calculation of the surface tension, it is the mass of the sample which is required and not the density, which may be subject to an uncertainty of ±5% [2]. The mass of the sample, on the other hand, can be easily measured to a much higher accuracy. Thus, the measurement accuracy of this method should be higher than that of the other methods involving the density.

In addition, this method is contamination free since the sample does not come into contact with the container. At high temperatures, when materials come in contact with a crucible, there is usually a chemical reaction at the interface and the material becomes contaminated. Furthermore, using this method, one can measure the surface tension over a wider temperature range since supercooling below the melting point frequently occurs because there are no nucleation sites for the solidification and at high temperatures, there are no reactions with the crucible. Thus, this method is very effective for measuring the surface tension of high temperature materials.

A sample is placed in the middle of the coil. When a high frequency alternating current flows through the coils, a magnetic field is produced, and an eddy current is generated on the sample's surface. Consequently, the electromagnetic force lifts the sample against gravity. Furthermore, the sample is heated and melted by the eddy current produced.

Although this method has many advantages as already mentioned, there is one disadvantage. When one tries to measure the surface tension under terrestrial conditions, the shape of the molten sample is distorted from a sphere, and consequently, there is splitting and a shift in the frequency of the surface oscillations of the droplet. Typically, there are five peaks in the frequency spectrum. Therefore,

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Rayleigh's equation cannot be used, and some corrections are necessary for the calculation of the surface tension. Although correction formulae have been proposed by Cummings and Blackburn [3] and by Suryanarayana and Bayazitoglu [4], the reliability of these correction procedures is still unclear.

To overcome this disadvantage, microgravity conditions have been used in this study. Microgravity conditions were obtained using the drop-shaft type system at the Japan Microgravity Center, JAMIC. In addition, a new method is proposed to enable minor changes to the droplet shape to be produced under microgravity.

2. EXPERIMENTAL PROCEDURE

Figure 1 shows a schematic diagram of the electromagnetic levitation apparatus. The size of the apparatus is about 1 m^3. As shown in this figure, the apparatus is equipped with a radio frequency generator, batteries, a pyrometer, high-speed video cameras, coils, and an infrared radiation heater. The radio frequency generator can supply a 14 kW a.c. current at 400 kHz for the positioning coil together with 1 MHz for the heating coil, as will be explained later.

In this study, the samples were 9 N pure silicon produced using the floating zone method. They weighed approximately 0.7 g. Because silicon has a high electric resistance at room temperature, an eddy current cannot flow in the sample. Therefore, in this study, an infrared radiation furnace was used for pre-heating the sample to a temperature where there is a significant decrease in the electric resistance. An atmosphere of Ar–3% H_2 was used, the gas being purified by platinum asbestos and magnesium perchlorate.

In order to obtain microgravity conditions, the drop-shaft type system at the Japan Microgravity Center, JAMIC, was used. The system has a 710 m deep hole including a braking zone to produce a 10 s microgravity of 10^{-5} G. The drop capsule is about 8 m long with a diameter of about 2 m, and the total weight is approximately 6 tons. Since the dropping hole is very long in this system, it cannot be evacuated. An air jet blows out from the nozzles located on the top of the capsule. In addition, the drop capsule consists of an inner capsule and an outer capsule in order to compensate for the air drag. A vacuum is maintained between the inner capsule and the outer capsule so that the free fall velocity of the inner capsule will not be affected by the air drag and the quality of microgravity is kept very high. Our experimental apparatus was placed in the inner capsule.

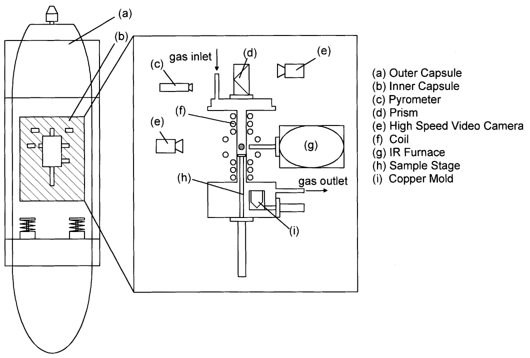

Fig. 1. Schematic diagram of the electromagnetic levitation system.

The experiment was carried out in the following way. At first, the sample was heated with the infrared radiation furnace, and then a high frequency current was generated in the coil. The sample was levitated, heated and melted by the eddy current. After the vertical and horizontal translation of the droplet decreased, the capsule drop was started. When the gravity changed from 1 to 10^{-5} G, the strength of the electromagnetic field was adjusted to stabilize the position of the droplet. A search coil was used to detect the motion of the molten silicon, and on the basis of this information, the coil current was adjusted to maintain the molten silicon in the middle of the coil.

The surface oscillation was recorded with two high-speed video cameras at 200 frames/s rather than using a photodiode, which has been widely used for the analysis of the frequency spectrum [5, 6]. The surface tension can be calculated from the surface oscillation frequency. When the free fall finished, the droplet was caught by a copper mould and the mass of the droplet was measured. The decrease in the mass during the experiments was less than 0.01%.

3. RESULTS AND DISCUSSION

3.1. Surface oscillation under microgravity

The shape changes of a levitated droplet are recorded using high-speed video cameras at 200 frames/s and the results are then subjected to a Fourier transformation to analyse the surface oscillation data. Figure 2 shows the Fourier transform of the change in the area of the droplet observed from the top under 1 and 10^{-5} G. The change in the area corresponds to the results obtained with a photodiode. Photodiodes have been widely used for the analysis of the surface oscillation data [5, 6]. As shown in the figure, the frequency spectrum contains quite a few peaks in the terrestrial condition. On the other hand, under microgravity condition only one main peak was observed. As one can see, microgravity conditions are very effective in simplifying the surface oscillations.

In this study, since the surface oscillations were monitored using a high-speed video camera rather

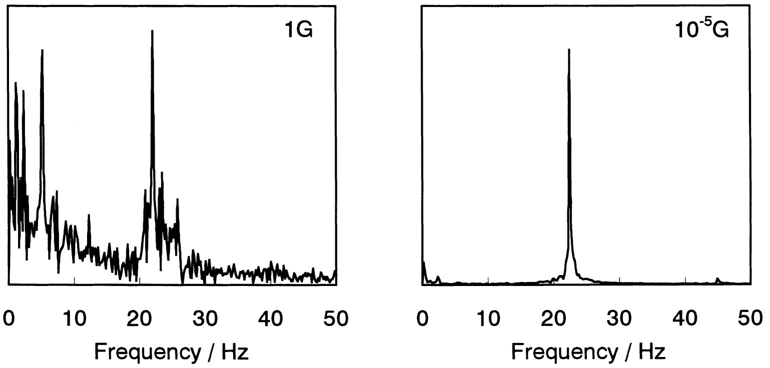

Fig. 2. Fourier transform of the change in area in 1 and 10^{-5} G.

than a photodiode, the surface oscillations can be analyzed in more detail [7]. For detailed analyses, data for the change in the radius of the droplet rather than the area were subjected to Fourier transformation. When the change in the radius is subjected to Fourier transformation, one peak is still observed in some cases, but in many other cases, there was splitting into several peaks, as shown in Fig. 3.

When the droplet is close to a sphere, five eigen oscillation frequencies can be generated for the oscillation of the $l = 2$ mode, as shown in Fig. 4. The various frequency peaks should correspond to these oscillations. When the droplet is a complete sphere, the frequencies of the five oscillation modes are the same and a single peak is observed. However, when the droplet is slightly distorted, the frequencies are different and several peaks are observed. When the

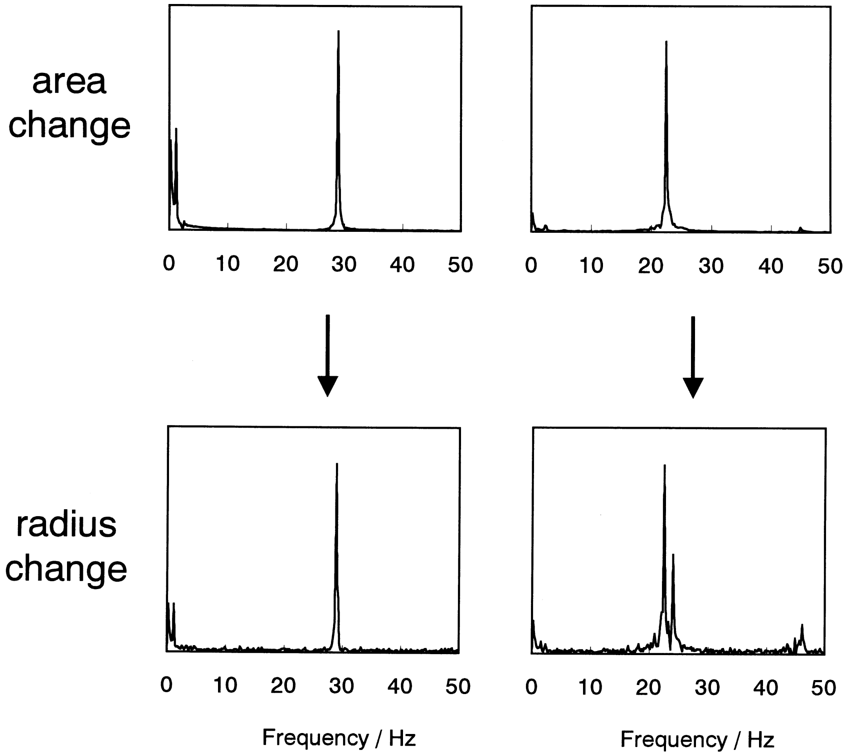

Fig. 3. Comparison of the Fourier transform of the change in area and radius of the top view. All oscillation modes do not give rise to an area change when the oscillation is observed from the top. Only when (a) the droplet is completely spherical, (b) the frequencies of the five oscillation modes are identical and (c) only one peak is observed in the radius change, can the oscillation frequency be attributed to the Rayleigh frequency.

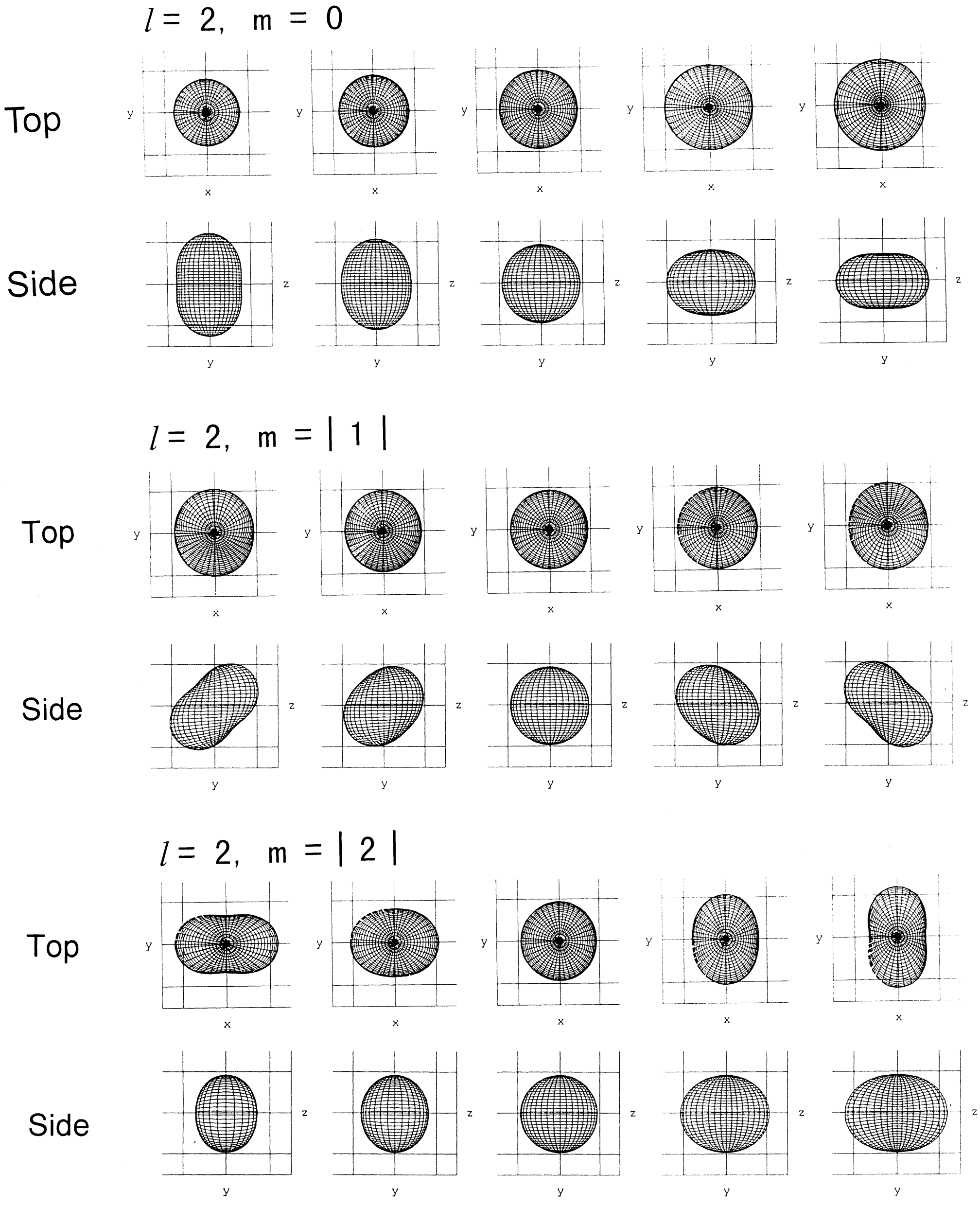

Fig. 4. Schematic view of five surface oscillation modes.

oscillations are observed from the top, not all oscillation modes give rise to an area change, as shown in Fig. 4. While the area change of the $l = 2$, $m = 0$ mode in the top view is very large, there is no area change for the $l = 2$, $m = |2|$ mode. Thus, it is essential to use the radius change in the analysis of the oscillations to confirm that there exists a single oscillation frequency.

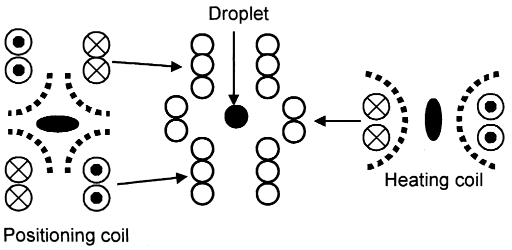

Fig. 5. Schematic diagram of the coil system.

3.2. Control of droplet shape

In this study, two types of coils were used, as

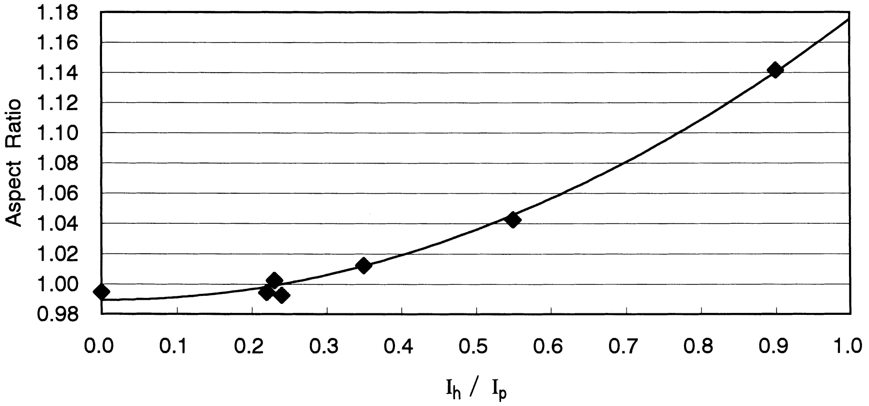

Fig. 6. Relationship between the aspect ratio of height/width of the side view and the ratio of coil current (heating coil current, I_h/positioning coil current, I_p).

shown in Fig. 5, in order to control the droplet shape. The positioning coil forms a quadrupole field that makes the droplet oblate. The heating coil forms a dipole field that makes the droplet prolate. Thus, the droplet shape can be changed by control of the current in the two coils.

Figure 6 shows the relationship between the droplet shape and the ratio of current for the heating coil, I_h/positioning coil, I_p. In this case, the current for the positioning coil was 100 or 200 A. As the current for the heating coil increases, the droplet shape becomes more prolate. When the current ratio is properly adjusted, the droplet shape can be a sphere, as shown in Fig. 7. The equilibrium shape was calculated by averaging 69 frames [8], because the droplet is in continuous vibration.

3.3. Relationship between droplet shape and peak distribution

As shown in Fig. 8, when the droplet shape is

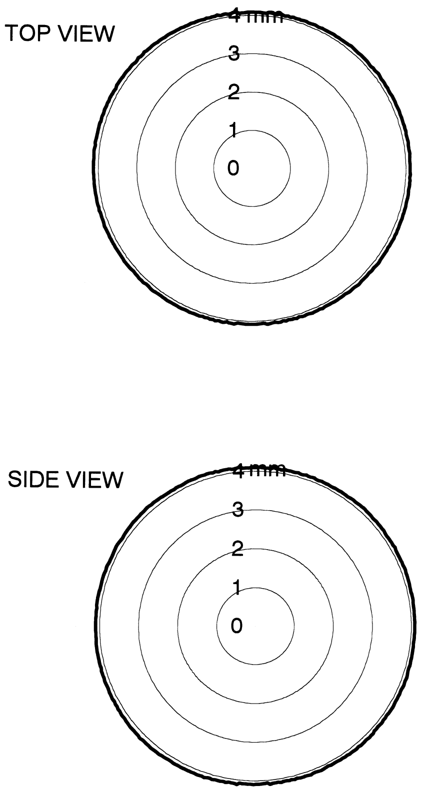

Fig. 7. Equilibrium shape of levitated droplet ($I_h/I_p = 0.22$).

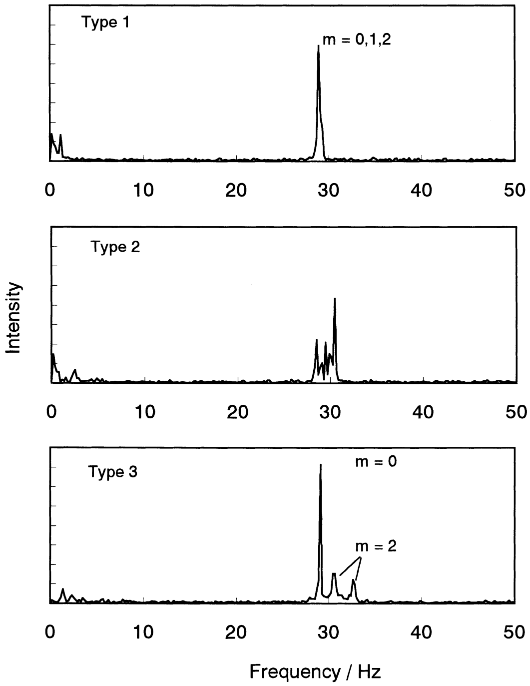

Fig. 8. Relationship between surface oscillation and equilibrium shape. (Type 1: spherical droplet, Type 2: slightly distorted droplet, Type 3: significantly distorted droplet.)

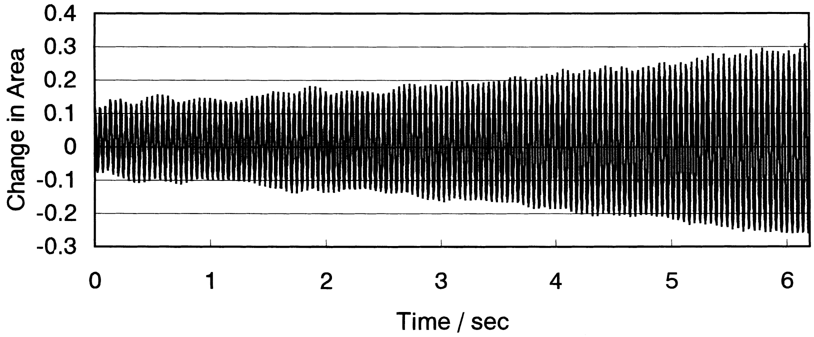

Fig. 9. Time dependence of surface oscillation when droplet is significantly distorted from sphere.

very close to a sphere, a single peak is detected in the oscillation spectrum. In this case, the oscillation frequencies of the five modes are the same, and consequently only one peak is detected. When the droplet shape is close to a sphere but is slightly distorted, five peaks are detected. Even a 1% distortion is enough to differentiate the oscillation frequency of the five modes. When the droplet is significantly distorted from a sphere, a very strong peak and some weak peaks are observed. For example, in the case of the results shown in Type 3 of Fig. 8, the strong peak corresponds to the oscillation of the $l = 2$, $m = 0$ mode and the weak peaks correspond to the oscillations of $l = 2$, $|m| = 2$. In this case, the amplitude of the $m = 0$ mode increases with time, as shown in Fig. 9. In this way,

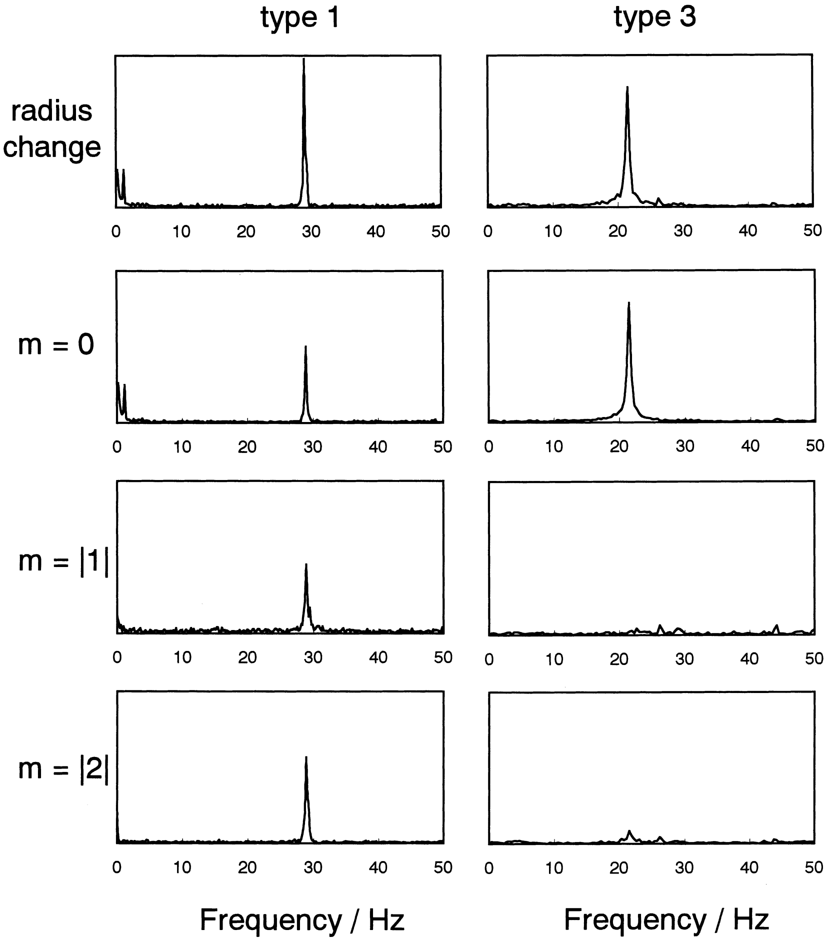

Fig. 10. Two types of Fourier transform with one peak. (Type 1: spherical droplet, Type3: significantly distorted droplet.)

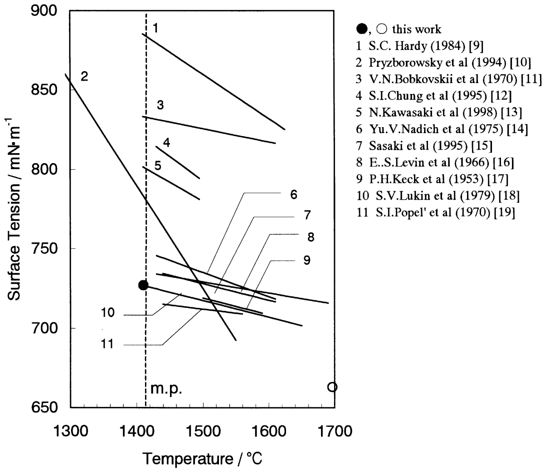

Fig. 11. Comparison of measured surface tension of molten silicon with values previously reported in the literature.

a particular mode is excited and the physical meaning of the weak peaks is not very clear.

This tendency is even clearer when a larger droplet is used. When a droplet twice the original size is used under the third condition, in some cases, only one peak is detected in the Fourier transform of the radius change, as shown in Fig. 10. In Type 1, only one peak is detected because the frequencies of the five modes are the same. In Type 3, however, only one peak exists since only one particular mode is detected.

When the peak in Type 1 is used to calculate the surface tension, the calculated value is close to the reported values [9–19], as shown in Fig. 11. The value of the surface tension obtained is 727 mN/m at 1683 K. However, when the peak in Type 3 is used, the calculated surface tension value is far from the reported values. Thus, when a single peak is obtained in the Fourier transform of the change in the radius of the droplet, one has to confirm that those five modes are included. In particular, it seems easy to obtain one peak when a particular pulse mode is given to the droplet, and therefore, one needs to confirm the existence of five peaks in this case to obtain non-scattered values of surface tension.

4. CONCLUSIONS

The effect of the shape change of a droplet on the surface oscillation has been investigated using the electromagnetic levitation method under microgravity. The following points have been established in this study.

1. Under the 10^{-5} G condition, the surface oscillation of a droplet is much simpler than that under 1 G.
2. The droplet shape is easily controlled by changing the current ratio between the quadrupole coil and the dipole coil, and a spherical shape can be obtained in this way.
3. When the droplet shape is spherical, only one peak is observed; when the droplet shape is close to a sphere, but slightly distorted, five peaks are observed. When the droplet is significantly distorted, a particular oscillation mode is usually excited and can result in a frequency spectrum with a single peak.
4. In order to obtain the correct surface tension value when only one peak is obtained, it must be confirmed that five oscillation modes exist and that all the oscillation frequencies are identical.

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