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Non-contact measurement of surface oscillation due to Marangoni flow instability in a silicon liquid bridge by phase-shift interferometry

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In order to investigate Marangoni flow instability of molten silicon, surface oscillation of a silicon liquid bridge with various aspect ratios at high Marangoni numbers, such as $Ma \geq 2400$, was observed by using real-time phase-shift interferometry. By analyzing phase distribution of the phase-shift interferograms using FFT (fast Fourier transformation) and wavelet transformation, we found that two frequency bands exist in surface oscillation. Their central frequencies are 0.1–0.5 Hz for a lower band and 0.7–1.3 Hz for a higher band, respectively. Central frequency decreases with increase in aspect ratio. The lower frequency bands, which include $m = 1$ and $m = 3$ modes, appear continuously, whereas the higher frequency bands appear intermittently.

1. Introduction

When difference of temperature and/or solute concentration exists on free surface, difference in surface tension occurs. Convection driven by these differences of surface tension is called Marangoni flow. The oscillatory Marangoni flow, which can be explicitly observed in microgravity conditions, causes defects in crystals, such as growth striation for instance. The non-dimensional Marangoni number used to scale the magnitude of Marangoni flow is defined as $Ma = -\sigma_T \Delta T h/\nu\rho\alpha$, where $\sigma_T = \partial\gamma/\partial T$; the temperature coefficient of surface tension, ΔT; the temperature difference of the liquid bridge, h; the characteristic length of the liquid bridge, ν; the kinematic viscosity, ρ; the density, α; thermal diffusivity. The Marangoni flow becomes stronger with increase in the Marangoni number, and consequently the flow is converted from a steady one to an oscillatory one with a single frequency and further to an oscillatory one with multiple frequencies.

For molten silicon, the Marangoni number is as high as $Ma = 1000$–10000 both under the float zone crystal growth condition and half-zone configuration. For the low-Pr number fluids, such as molten silicon, it has been reported by numerical and experimental studies that the azimuthal mode wave number m due to flow instability depends on the aspect ratio of the half-zone bridge: $m{\cdot}As \approx 2$ [1–3]. Here, the aspect ratio As is defined as the ratio of the bridge height h to the radius r. Concerning frequency of the instability mode for a high Pr-number fluid, Preisser *et al.* reported that frequency decreases with increase in the aspect ratio [4]. Ueno *et al.* observed experimentally evolution of oscillation mode with increase in the Marangoni number for a high Pr-number fluid [5]. However, for low Pr-number fluids, the relationship between the frequency and the aspect ratio has not been experimentally investigated, although the oscillation frequency was calculated for the specific azimuthal wave number m for the low Marangoni number case [6]. For the high Marangoni number case, such as $Ma = 1000$–10000, instability of Marangoni flow would develop to a significant level; i.e., flow would become three dimensional and time-dependent. However, through temperature oscillation measurement, it was estimated that the basic structure of flow instability, which can be explicitly observed for the low Marangoni number case, was still sustained even for such a high Marangoni number case: $m{\cdot}As \approx 2$ [3]. In such a condition, however, a clear single peak was not observed but frequencies were continuous and multiple; i.e., frequency bands were observed. In order to study oscillatory behavior more in detail and clearly, it was suggested to decrease the Marangoni number.

When flow instability takes place, temperature, flow and pressure fields become non-axisymmetric and oscillatory in the liquid bridge [1, 6]. Due to oscillation of the pressure fields within the liquid bridge, oscillation takes place also at the surface of the liquid bridge. Therefore, it is conceivable that observation of the surface oscillation enables us to investigate the Marangoni flow instability in the liquid bridge [7, 8]. Surface oscillation observation could overcome limitations caused by the time and spatial resolutions of temperature oscillation measurement using thermocouples. Also, insertion of thermocouples could contaminate the liquid system and interfere with the flow field.

In the present study, for a molten silicon bridge with a half-zone structure with Marangoni number of $Ma = 2400–11200$, we observed surface oscillation using real-time phase-shift interferometry. The phase distribution of interferogram was analyzed by FFT (fast Fourier transformation) and wavelet transformation [9] techniques to clarify the frequency of surface oscillation and its time dependence. The characteristics of oscillation, i.e. continuous or intermittent, and the relationship between central frequency of the frequency band and the aspect ratio are discussed.

2. Experiment and data analysis

2.1. Experimental setup

We used a mirror furnace to prepare a half-zone molten silicon bridge. A 1300 W halogen lamp, positioned at the upper focus point, served as a heat source. A quartz ampoule containing a silicon sample was placed at the lower focus point [2]. The diameter of the liquid bridge was 10 mm and the height was 3.0, 5.0, 7.0 and 10.0 mm; the corresponding aspect ratios *As* were 0.6, 1.0, 1.4 and 2.0. The molten silicon bridge was fixed between the upper and the lower rods made of carbon. For this configuration, the Marangoni number was estimated to be 3000 to 14000 depending on the aspect ratio [3]. In order to decrease the Marangoni number and to observe the frequency of the flow instability more easily, a hybrid rod was prepared for the lower one. The hybrid rod consisted of an alumina rod, and a short carbon rod that was inserted between the alumina rod and molten silicon. An alumina rod with smaller thermal conductivity than that of carbon was used to decrease heat transfer from the molten silicon to the main body of the mirror furnace through the lower rod, so that temperature difference between the upper and lower interfaces of molten silicon could be decreased. A short carbon rod was attached to the alumina rod to assure wettability of molten silicon with the lower rod. In order to confirm this effect, we prepared a molten silicon bridge, as the temperature of the melt at the lower interface was set be the melting point of silicon. Under this condition, heat flux applied to upper rod was decreased 20%, compared to the conventional case where carbon rods were used both for the upper and lower rods. Consequently the Marangoni number was decreased by 20% down to $Ma = 2400–11200$.

As shown in Fig. 1, we employed a real time phase-shift Michelson interferometer. The optical system was installed on the vibration-isolated bench to eliminate the oscillation induced by external sources over 2–3 Hz [7]. An Ar-laser with a wavelength of 488 nm was used as a light source for the interferometer. By using this wavelength, it is easy to distinguish the light of interference from radiation from the silicon melt surface, which contains long wavelength components. Irradiating both the silicon melt surface and the reference solid silicon rod surface by Ar laser beam, interference took place due to optical path difference. The interferograms were recorded by a high-speed camera "Motion Scope PCI 1000s". Depending on the frequency of surface oscillation, sampling rate was optimized. Due to characteristics of the wavelet transformation, the higher the observed frequency becomes, the shorter becomes the duration for observation. Thus, ambiguity is high for the high frequency region, when the sampling rate is low. This is also the case for the low frequency region when the sampling rate is high because of the limit on recording time. Therefore, we observed oscillation of the high frequency region (>0.5 Hz) at sampling rates of 250 and 500 fps and of low frequency region (<0.5 Hz) at sampling rates of 60 and 125 fps.

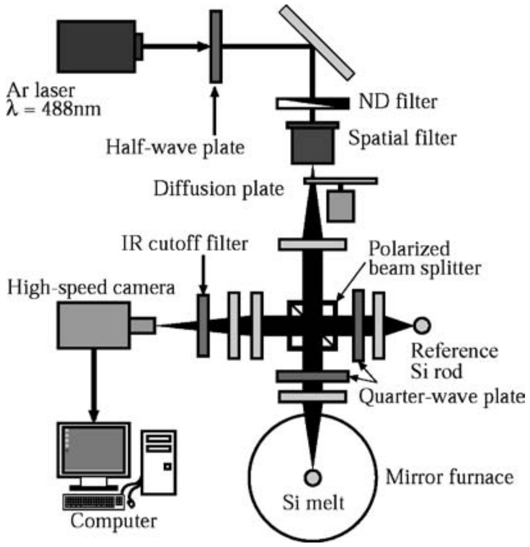


Figure 1 Setup for phase-shift Michelson interferometer.

2.2. Data analysis; using of phase-shift-interferometry

In order to observe surface oscillation of molten silicon bridge precisely, we applied a single-camera phase-shift-technique to the Michelson interferometer. Examples for utilization of this technique to surface oscillation analysis were reported elsewhere [7, 8]. Height distribution of the observed plane, which corresponds to distribution of radius due to oscillation, was obtained by analyzing information of the phase distribution with improved spatial resolution. If the height distribution can be observed dynamically, oscillation of the observed plane can be analyzed. The limit of spatial resolution of conventional optical interferometry is 1/4 of a wavelength, whereas that of the phase-shift interferometry is 1/512 of a wavelength. Since we used an Ar-laser with a wavelength of 488 nm in the present investigation, the spatial resolution was almost 1nm. We can know dynamic deformation of the surface of the liquid bridge, through analyzing the radial displacement (RD), azimuthal phase value gradient (AZ) and axial phase value gradient (AX). Consequently we can obtain the frequency of the surface oscillation by using FFT and wavelet transformation. Wavelet transformation is one of convenient methods to analyze time evolution of oscillation frequency [9], whereas FFT is a popular method which can provide spectrum distribution with high accuracy. However, time-dependent information might be lost for the FFT, when long time span is concerned. Using the Gabor function as a mother wavelet, we prepared a signal plane to visually analyze time evolution of oscillation frequency for liquid bridges with various aspect ratios.

3. Results and discussion

For a highly developed oscillatory flow, it is difficult to determine the frequency by using FFT, because peaks of the frequency are multiple, continuous and time-dependent. Instead, we used a wavelet transformation technique and prepared a signal plane, so as to confirm that there is no single, sharp, peak and that there are frequency bands which show time-dependent behavior. As a result of the signal plane and FFT analyses, we extracted the basic structure of the flow instability in the liquid bridge for each aspect ratio.

3.1. Higher Marangoni number case (use of carbon rod)

Fig. 2 shows a signal plane for frequency of surface oscillation (radial displacement) in the liquid bridge with $Ma \approx 7000$ and $As = 1.2$; the sampling rate was 500 fps. When strong black contrast appears in the signal plane, this suggests that the amplitude of oscillation is strong. As shown in Fig. 2, a band, whose central frequency is about 0.5 Hz, likely exists continuously from $t = 3.0$ s on. There is an unsteady frequency band whose central frequency appears at $f = 1.2$ Hz and above; oscillation takes place intermittently. The results of FFT analysis show that there were clear peaks at around $f = 0.5$ Hz, but near $f = 1.2$ Hz and above there were no clear peaks of frequency but peaks were distributed continuously, as shown in Fig. 3. From these experimental results, we assume that frequency band, whose central frequency was 0.5 Hz and less, corresponds to that of basic structure of Marangoni flow instability in the half-zone liquid bride; and the frequency that appeared at 1.2 Hz and higher could be generated by formation of small vortices due to high Marangoni number. A similar tendency was observed, regardless of aspect ratio As and sampling rate; we could not observe aspect ratio dependence of flow instability structure. This is probably due to too high Marangoni number of 3000–14000.

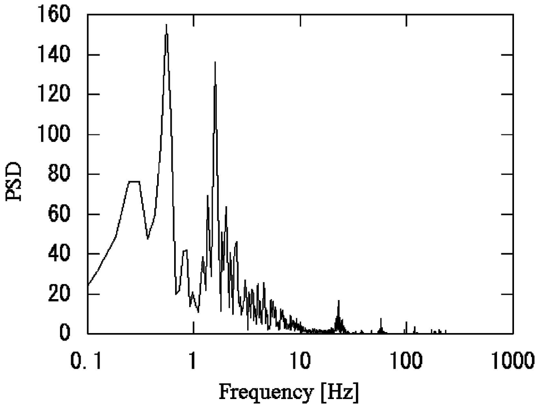


Figure 3 Power spectrum density of the FFT analysis for the same configuration as that shown in Fig. 2.

3.2. Reduced Marangoni number case (use of hybrid rod)

In order to observe the basic structure of the flow instability in the half-zone structure more accurately, we prepared half-zone bridges with a relatively low Marangoni number. This was attained by employing a lower rod with hybrid structure with an alumina rod and a carbon attachment with larger thermal resistance than that made of carbon. By employing this configuration, the Marangoni number was estimated to be 20% smaller than that for the conventional configuration which utilizes carbon rods both for the upper and lower rods. The Marangoni number for this configuration was estimated to be 2400–11000 depending on the aspect ratio.

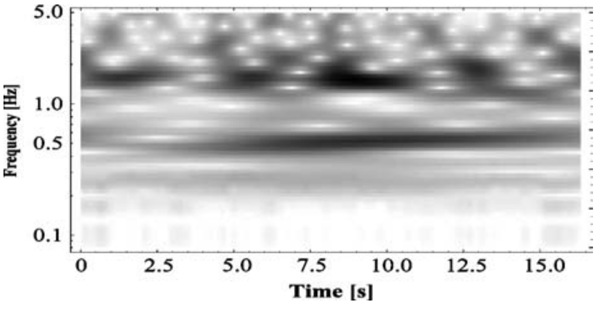
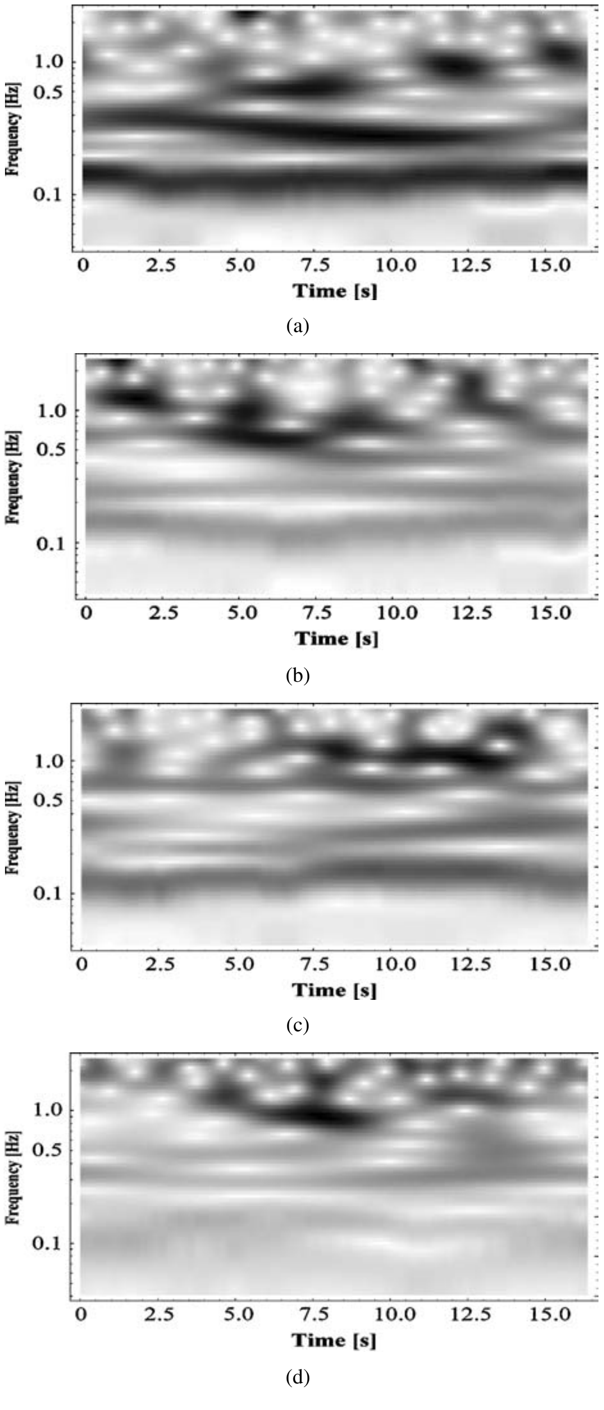


Figure 2 The signal plane obtained by wavelet transformation using Gabor function, which shows oscillation of radial displacement (RD) for the molten silicon bridge with an aspect ratio of $As = 1.2$ and Marangoni number of $Ma \approx 7000$.

The results of the frequency plane analyses for the molten silicon bridges with reduced Marangoni numbers are shown in Fig. 4a–d; Fig. 4a and b show oscillation of axial phase value gradient (AX) and Fig. 4c and d show oscillation of radial displacement (RD). Frequency band structure was not easily observed when data was analyzed using oscillation of azimuthal phase value gradient (AZ); this is probably due to smaller amplitude of AZ compared with that of RD and AX.

It is clear that the frequency bands were observed more explicitly compared with that for the higher Marangoni number case (Fig. 2). This is due to stabilization of flow at small Marangoni number, which makes oscillation frequencies more distinguishable. Frequency is not a single line but has some width; this suggests that a frequency is no more single one but appears to fluctuate around the central frequency. For the case shown in Fig. 4a ($As = 0.6$, $Ma \approx 2400$), fluctuation width was 0.05 Hz for the frequency band with the central frequency was 0.14 Hz. Frequency band structures with the central frequency less than 0.5 Hz appear all over the experimental duration, whereas time-dependent behavior appeared above 0.5 Hz. As shown in Figs 4a–d, amplitude of frequency, i.e. black contrast of frequency plane, was strong for the liquid columns with small aspect ratio and small Marangoni number, and frequency with the strongest amplitude moves to higher frequency with increase in aspect ratio. This suggests that small but strong vortices were formed with increase in the Marangoni number; in the present experiment the Marangoni number increases with increase in aspect ratio.

Fig. 5 summarizes the preliminary results of wavelet analysis for surface oscillation as a function of an



(a)

(b)

(c)

(d)

Figure 4 The signal plane obtained for the molten silicon bridges with various aspect ratios and reduced Marangoni numbers attained by use of a hybrid lower rod: (a) $As = 0.6$, $Ma \approx 2400$, (b) $As = 1.0$, $Ma \approx 4000$, (c) $As = 1.4$, $Ma \approx 6800$, (d) As $= 2.0$, $Ma \approx 11200$. Fig. 4a and b show oscillation of axial phase value gradient (AX) and Fig. 4c and b show oscillation of oscillation of radial displacement (RD).

aspect ratio, which are shown in Fig. 4a–d. As shown in Fig. 5, there are likely two groups of frequency bands. It is noteworthy that the lower frequency bands, whose central frequency ranges from 0.1 to 0.35 Hz, were distributed over the whole duration of observation, whereas high frequency bands whose central frequency ranged from 0.5 to 1.3 Hz, was distributed in a less orderly way; i.e., oscillation was observed intermittently and frequency ranged wider. According to dynamic temperature oscillation analysis, frequency of the $m = 3$ mode was found to range from 0.01 to 0.2 Hz and that of the $m = 1$ mode from 0.08 to 0.2 Hz [3]. This suggests that lower frequency band in Fig. 5 consists not only of a single mode but also of multiple modes, such as mixture of $m = 1$ and $m = 3$ modes. Furthermore, each mode would be widened. This is supported by appearance of two or three bands below 0.5 Hz in a frequency plane, as shown in Fig. 4a–d. Furthermore, temperature oscillation measurement also reports that the appearance ratio of each mode shows aspect ratio dependence. Higher frequency bands whose central frequency ranged from 0.5 to 1.3 Hz appear intermittently. Furthermore, oscillation with much higher frequency than that was observed. These are attributed to generation of small but strong vortices due to high Marangoni number.

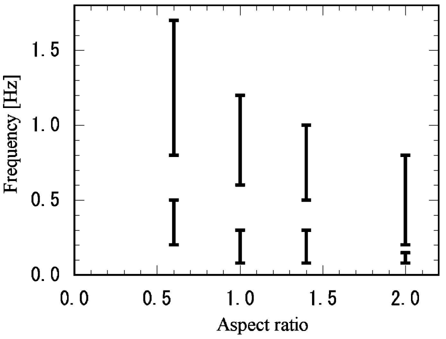


Figure 5 The relationship between the frequency of the surface oscillation and the aspect ratio *As*. The lower bands appear continuously, whereas the higher band appear intermittently.

Fig. 5 shows the relationship between frequency of surface oscillation and aspect ratio *As*. As shown in Fig. 5, it is clear that the frequency of surface oscillation decreases with increases in the aspect ratio *As*. This tendency agrees with that observed for high Pr-number fluid reported by Preisser *et al.* [4]. For their experiment, only the aspect ratio changed keeping the Marangoni number constant, whereas both Marangoni number and aspect ratio were changed simultaneously in the present investigation, because it is experimentally difficult to control both the melt height and the temperature difference between the upper and lower interfaces independently. For high Pr-number fluids, the frequency of basic instability structure almost does not depend on the Marangoni number [5]. We can conclude from the present study that the frequency depends more on aspect ratio than on the Marangoni number; however, there still remains the possibility that the frequency of each basic instability mode changes with changes in the Marangoni numbers. In order to distinguish the effect of the Marangoni number on frequency from that of aspect ratio more clearly, the more precise experiments are required. Also, studies, which can bridge physics obtained numerically at the low Marangoni number case and that obtained experimentally at the high Marangoni number case, are necessary to understand clearly instability of Marangoni flow of low Pr-number fluids. Simultaneous observation of both temperature oscillation and surface oscillation could clarify the relationship between frequency, azimuthal mode and aspect ratio.

4. Summary

Surface oscillation due to Marangoni flow instability in a silicon liquid bridge with various aspect ratios ($As = 0.6–2.0$) was investigated by using real-time phase-shift interferometry. Through analyzing the frequency of surface oscillation using the FFT and wavelet transformation analyses, the followings are clarified;

- From the FFT and wavelet transformation analyses, two frequency bands were identified. The lower one, whose central frequency ranges from 0.1 to 0.35 Hz, was distributed over the whole duration of the experiment, whereas high frequency bands whose central frequency ranged from 0.5 to 1.3 H appeared intermittently.
- Comparing the present results with that obtained by temperature oscillation measurement, the lower band would consist of multiple modes, such as $m = 1$ and $m = 3$.
- Frequency for surface oscillation decreased with increasing aspect ratio As. This tendency agrees with that for high Pr-number fluids observed by Preisser *et al*. For low Pr-number fluids, frequency appears to depend rather on As than on Marangoni number.
- Additional studies, which can bridge physics obtained numerically at the low Marangoni number case and that obtained experimentally at the high Marangoni number case, are necessary to understand clearly instability of Marangoni flow of low Pr-number fluids.

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