Journal of Crystal Growth 318 (2011) 156–161

Contents lists available at ScienceDirect

Journal of Crystal Growth

journal homepage: www.elsevier.com/locate/jcrysgro

On experimental and numerical prediction of instabilities in Czochralski melt flow configuration

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ARTICLE INFO

Available online 2 November 2010

Keywords:
A1. Fluid flows
A1. Convection
A1. Marangoni convection
A1. Thermocapillary convection
A2. Czochralski method

ABSTRACT

A new experimental facility for experimental validation of numerical codes calculating Czochralski melt flow is described. The setup is built to make all the boundary conditions reproducible in a mathematical model. Measurements are focused on the steady-oscillatory flow transition, which is defined by appearance of temperature oscillations measured independently by thermocouples and interferometer. Location of thermocouples is defined according to numerical predictions, which ensures measurements at locations where oscillations amplitude is large. Current state of comparison of experimental and numerical results is discussed.

1. Introduction

In this study we address a problem of validation of numerical codes and methods applied for various problems of crystal growth. An unprecedented growth of computational power during last three decades turned computational modeling from a kind of art to a standard and widely used research instrument. Apparently, like every other research tool, each computational model has its level of accuracy and tolerance, which must be known to make the whole research reliable. This poses non-trivial problems of estimation of numerical error, verification, and validation of the computational codes. Methods of estimation of numerical error (see, e.g., [1]) are being developed, however they did not reach yet the complicacy of crystal growth modeling. According to a recently adopted terminology [2], "verification" means comparison of results obtained by independent codes. This task is commonly recognized, however proposed benchmark tests related to crystal growth, e.g. [3–5], did not attract too much attention. The term "validation" of numerical codes relates to comparison of computational results with laboratory experiment. The importance of such comparisons is unquestionable, however it requires experiments to be carried out with numerically (or mathematically) reproducible initial and boundary conditions. Otherwise, quantitative comparison of experimental and numerical results is not possible. It seems that the latter characterizes also present situation in crystal growth computational modeling: numerical results yield mainly qualitative descriptions, while numerous experiments are not defined or described with enough details to be accurately reproduced in computational models to make the quantitative comparison possible.

In this study we focus on an experimental hydrodynamic model of Czochralski melt flow, which is similar to one used in [6–8], but whose thermal boundary conditions are defined more precisely. Temperature measurements are done by thermocouples and an interferometer, which cross-verify each other and strengthen our confidence in the measured results. We focus on steady-oscillatory and/or axisymmetry-breaking transitions of the flow by measuring critical temperature difference ΔT and appearing frequency of oscillations. Then we measure how the frequency spectrum changes with an increase in ΔT. These results, especially those related to the instability threshold, are to be compared with computational simulation. We assume that quantitative agreement between measured and calculated critical parameters, which is yet to be obtained, ensures correct calculation of the flow and temperature fields, at least not very far from the threshold. This assumption is based on several conclusions derived from computational stability studies [9,10], where it was stated that correct computation of the instability threshold requires a sufficient accuracy for computation of both steady-state flow and the leading unstable eigenmode. On this basis all additional information can be derived from the numerical solution without carrying out new experiments. In particular, such approach allows one to avoid complicated and expensive experimental visualizations of the flow and temperature fields.

2. Experimental setup

Special efforts were made to improve the sensitivity of temperature fluctuations measurements by both Mach-Zehnder interferometer and thermocouples. In the interferometry measurements the phase lag is integrated along a laser beam, so that only its coherent part can be

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doi:10.1016/j.jcrysgro.2010.10.013

revealed. To avoid refraction effects the beam passes through the system axis of symmetry. Obviously, the phase lag strongly depends on the type of measured disturbance. Thus, according to the linear stability theory small perturbations appear as azimuthal Fourier modes, i.e., $\sim\exp(im\theta)$. Then the even modes ($m=0$, 2, 4, etc.) are symmetric for rotation in 180° around the apparatus axis and are expected to provide notable phase lags. At the same time, odd modes are antisymmetric and may result in nearly zero lag. Careful alignment of the interferometer and use of optical filters enabled us to obtain very distinct and clear fringes at the focal point where the distorted and undistorted rays cross each other. These fringes fluctuate when oscillating perturbation (e.g., cold plumes detaching from the dummy crystal [8]) develops in the vicinity of the axis. Very high sensitivity and precision of temperature measurements by the thermocouples are achieved by employing high-resolution data acquisition system, which includes a low-noise amplifier and 16 bit-A/D converter, which are calibrated in situ. The calibration is carried out by creating steady motionless conditions within the crucible keeping the same temperature at the crucible boundaries and the crystal dummy. The resulting temperature is measured by a platinum thermometer whose precision is better than 0.01 °C. It should be noted that the calibration and actual measurements are extremely time consuming. An average experimental run needs about 5 h for the calibration and 8 h for the measurements at each new condition. In this series of experiments we vary temperature difference and angular velocity of the dummy. A special program in LABVIEW was developed to enable fully automatic run of the experiments.

The experimental setup is sketched in Fig. 1. It consists of precise 90 mm diameter cylindrical container (1) made from transparent glass and a 45 mm cold copper dummy (2) imitating a crystal pulled out from the melt in a crystal growth facility. The latter plays a role of a cold rotating cover. The glass container is put inside a doubleglass envelope with vacuum between its walls (5). The crucible and the envelope are closed by Teflon covers with an O-ring (3). Isothermal water, supplied by a computer controlled thermal bath is flowing between the envelope and the crucible. The doubleglass envelope and the Teflon covers ensure a very good thermal insulation, so that after a certain time period the container wall and bottom become perfectly isothermal.

The copper dummy is cooled by the cold flowing water supplied by another computer-controlled thermal bath. To ensure an intensive cooling and a constant temperature on its surface the dummy is hollow and has copper ribs inside it, so that heat transfer between the dummy lower surface and the cold water is enhanced. Electric brushless DC motor is installed to enable the dummy rotation. A suitable control unit allows for adjustment of rotation rate in the range of 0.001–1 revolutions per second according to experimental needs. Temperature fluctuations are measured by very fine 0.1 mm diameter, T-type thermocouple wires (6). One pair of the thermocouples is placed at the corner of the crucible and another pair – under crystal dummy near to crucible bottom. The interferometry setup consists of two beam splitters (7), two mirrors (8), and beam expander (9) that allow for a precise adjustment of the laser beams to obtain high-quality interferometry fringes on the screen (10). The fringes are filmed by a CCD camera (11). All signals from thermocouples and camera are transmitted to PC for post-processing.

In the experiments reported here the experimental liquid was distilled water whose Prandtl number ($Pr\approx 6.6$) is close to that of oxide melts [5]. The crucible was filled up to the aspect ratio height/radius $=1$. Water was chosen as the simplest possible experimental liquid that allowed us to finalize the design of our setup. We are aware of the fact that water can be a problematic choice because its parameters are strongly temperature dependent and its large surface tension causes contamination of the interface. To overcome the first difficulty we made the size of our setup rather large, so that the Grashof and Marangoni numbers are estimated as $Gr\approx 222{,}000\Delta T$

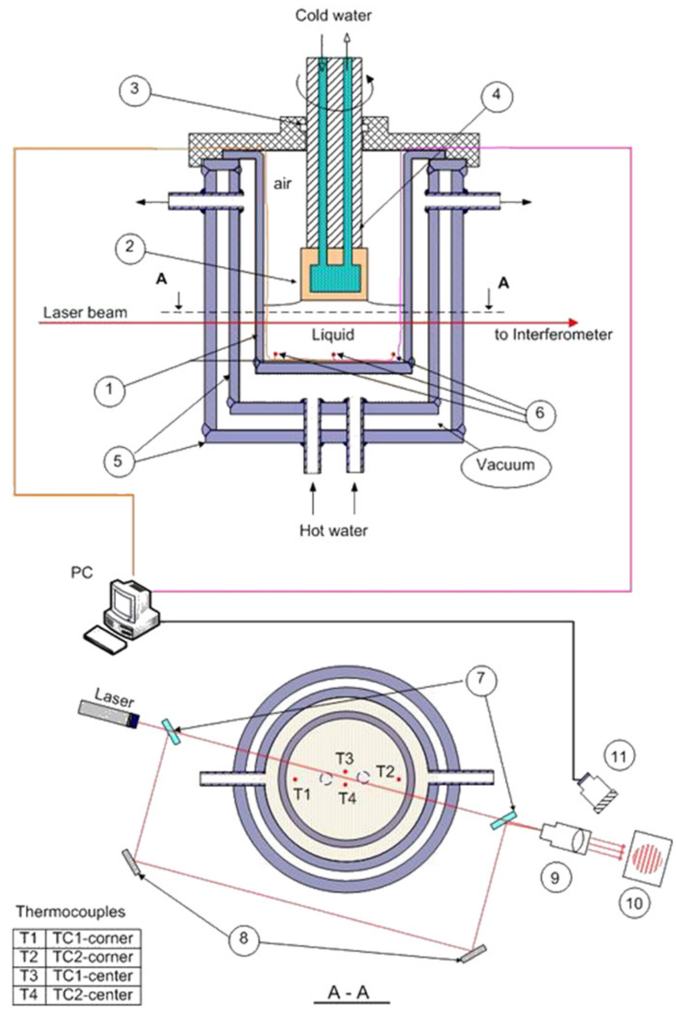

Fig. 1. Sketch of experimental setup. Explanations are given in the text.

and $Ma\approx 50{,}000\Delta T$, respectively, which makes the critical temperature difference smaller than 1 °C. This allows us to neglect temperature dependence of water thermophysical properties when computing the instability threshold. The contamination of free surface is more difficult issue that possibly alters the flow in a way that cannot be reproduced in our numerical model. We plan to address this issue in future studies, possibly by replacing the experimental liquid.

To obtain correct thermal conditions at the free surface we solve not only for the flow in the crucible, but also for convection of air above, which results in a large additional computational effort. We have found that postulating the Newton convection law and fixing a certain Biot number, as is done, e.g., in [5,6], alters the base flow and its stability properties too strongly. We are certain that a correct computation of heat transfer above the melt free surface is essential for reliable computational modeling of model experiments, as well as of the real CZ processes.

An example of calculated flow is shown in Fig. 2 together with the pattern of a characteristic temperature perturbation mode. The lower pattern of streamlines correspond to the working liquid, while the upper part – to to convection of air. Note a small circulation between two large vortices, which develops because the water and the air rise along the heated sidewall, so that main circulations of air and water near the free surface have opposite directions. This small circulation alters heat transfer from the free surface and has to be taken into account.

Pattern of the temperature perturbation exhibits local maxima where temperature oscillation amplitudes are expected to be the largest. Other perturbation modes also have maxima either below the cold dummy or in the corner, or in both places. This finding determined where to install the thermocouples. Really, in our early experiments the thermocouples were installed around the crystal just below the free surface. With such an installation it was not possible to measure instabilities reported below.

In the reported experiments two pairs of thermocouples were installed close to the apparatus axis, and two others at the crucible lower corner in accordance with the numerical predictions. A certain effort was made to locate each pair of thermocouples symmetrically relative to the apparatus axis. The pairs are shifted relatively to each other by 90°. The probes are glued to the side walls and the bottom and thus less obstruct the flow.

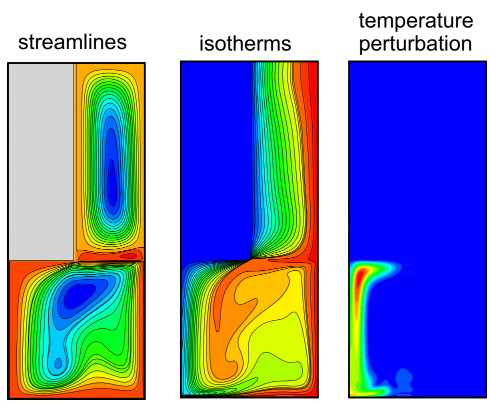

Fig. 2. Pattern (streamlines and isotherms) of calculated steady flow at $\Delta T = 1\,^\circ$C and absolute value of the most unstable temperature perturbation (right).

Table 1
Experimental parameters and measured fundamental frequencies and amplitudes.

Runs	n (rpm)	ΔT (°C)	TC-center		TC-corner	
			f (Hz)	Amplitude (°C)	f (Hz)	Amplitude (°C)
1	0	0	–	–	–	–
		0.1	–	–	–	–
		0.2	–	–	–	–
		0.4	0.021	0.0026	–	–
		0.5	0.023	0.0057	–	–
		0.6	0.026	0.0127	–	–
		0.7	0.028	0.0265	–	–
		0.8	0.030	0.0550	–	–
		0.9	0.032	0.0687	0.032	0.00072
		1	0.034	0.0732	0.034	0.00085
2	0	0	–	–	–	–
		1	0.034	0.08217	0.034	0.00048
		2	0.056	0.03633	0.056	0.002
		3	0.076	0.03292	–	–
		4	0.093	0.12919	–	–
		5	0.1	0.20298	–	–
		6	0.118	0.13267	–	–
		7	0.128	0.07782	–	–
		8	0.14	0.10954	–	–
3	1	0	–	–	–	–
		0.2	0.006	0.01342	–	–
		0.4	–	–	–	–
		0.6	0.003	0.00438	0.003	0.00764
		0.8	0.005	0.00587	0.005	0.00453
		1	0.007	0.00424	0.007	0.00849
4	4	0	–	–	–	–
		0.2	–	–	–	–
		0.4	0.008	0.00735	0.008	0.01386
		0.6	0.007	0.01123	0.007	0.02417
		0.8	0.008	0.00980	0.007	0.01875
		1	0.01	0.01249	0.008	0.01603

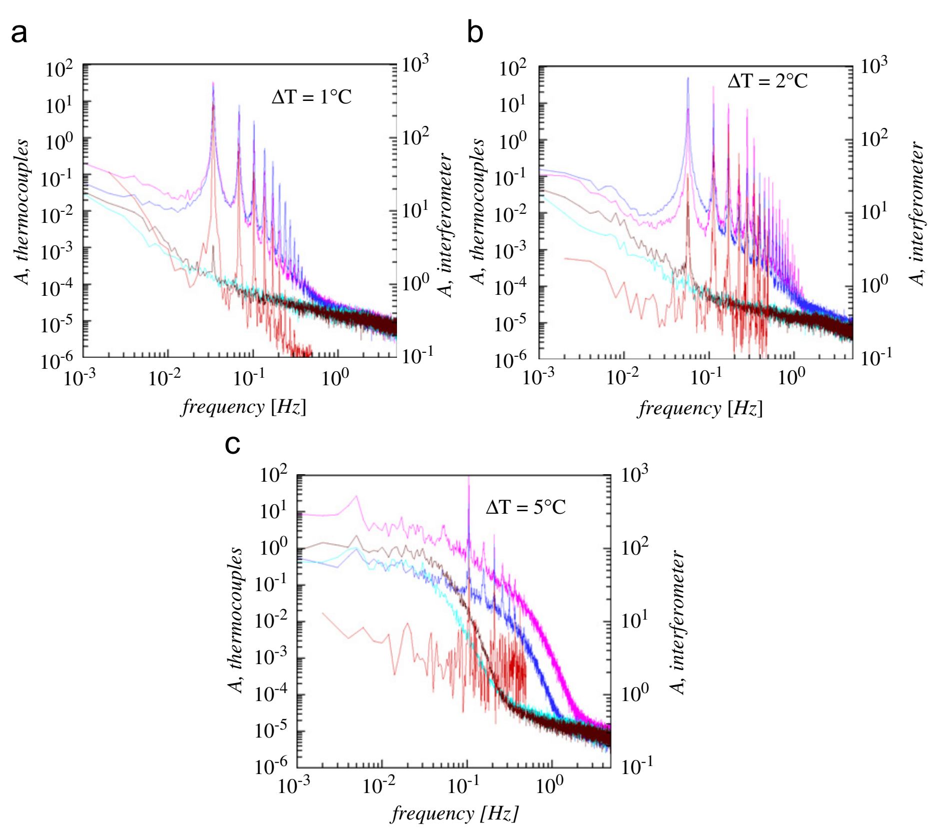

Fig. 3. Power spectrum measured at different temperature differences without dummy rotation. Red color corresponds to interferometer, pink and blue, to thermocouples placed in the center, black and cyan, to thermocouples placed in the corners. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

3. Results

Three sets of experiments were conducted with the dummy rotation at angular velocity 0, 1, and 4 rpm. The temperature difference was varied from 0 to 8 °C.

Examples of measured temperature oscillations spectra are shown in Fig. 3. Table 1 presents fundamental frequency of oscillations and its amplitude for thermocouples located near the apparatus axis and in the crucible corner for temperature differences and angular dummy velocities of main experimental runs. To ensure repeatability of results several experimental runs were performed at each parameter set.

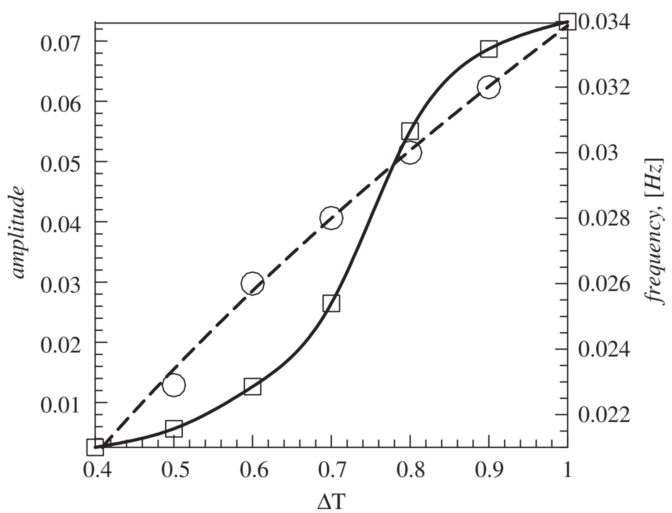

Fig. 4. Amplitude (squares and solid line) and frequency (circles and dash line) of the main harmonics versus temperature difference.

The first striking result is a perfect agreement between fundamental frequency and its harmonics measured by the interferometer and thermocouples located in the center of the apparatus (Fig. 3). The pair located in the corner indicates significantly lower amplitude values in accordance with the perturbation pattern shown in Fig. 2. We assume that this is the "cold plume" instability observed in [8], which is supported also by some preliminary flow visualizations (not reported here).

As follows from Table 1 in the case of stationary crystal the amplitude and the frequency monotonically grow with increase in the temperature difference, which is also illustrated in Figs. 4 and 5a. This allows us to assume that the instability sets in as a supercritical bifurcation, which is also an important indication for future computational modeling. The results in Fig. 5 are shown only for one thermocouple located in the crucible center, since amplitudes in the center are significantly larger than those in the corner. This tendency remains unchanged until some notable spectral density is detected by the corner thermocouple at relatively large temperature difference $\Delta T = 1$ °C.

In the case of a slow rotation of the crystal dummy ($\Omega = 1$ rpm, Table 1 and Fig. 5b) we observe appearance and disappearance of instability with the gradual increase in ΔT (Table 1). Possibly, this is due to a very complicated dependence of ΔT_{cr} on the crystal rotation, especially at low rotation rates, as was found in our numerical

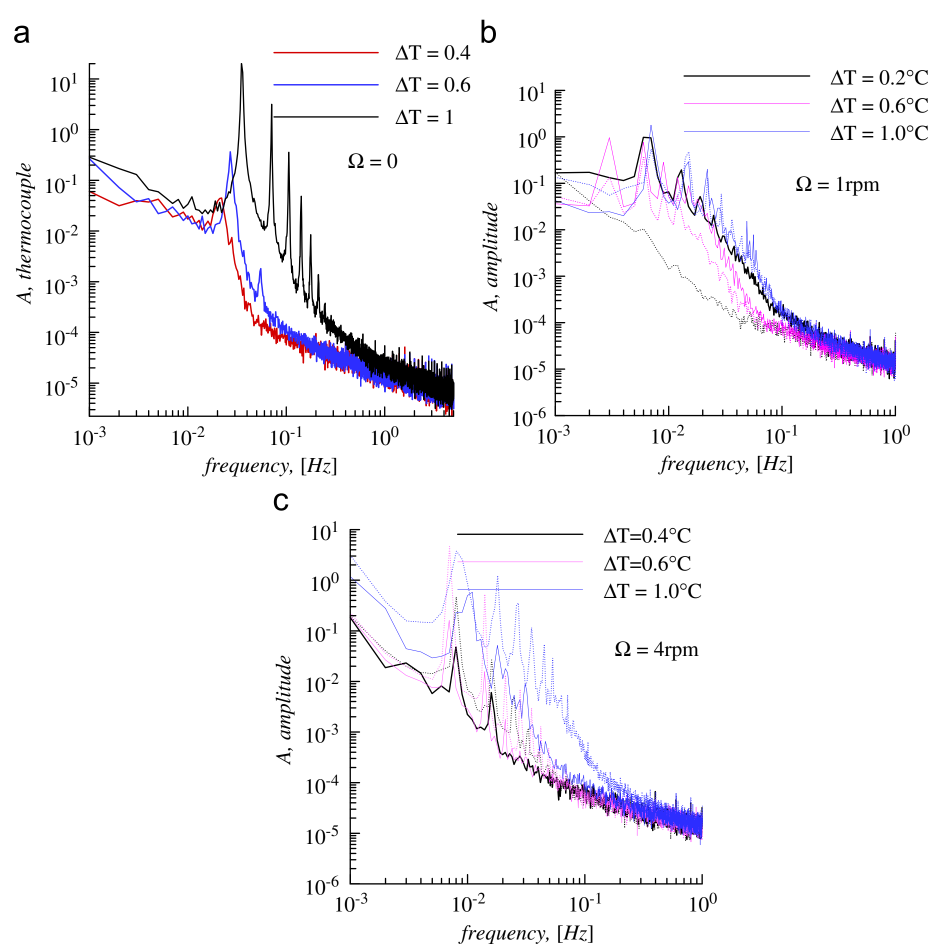

Fig. 5. Power spectrum measured at different temperature differences and at different dummy rotations by a thermocouple placed in the center.

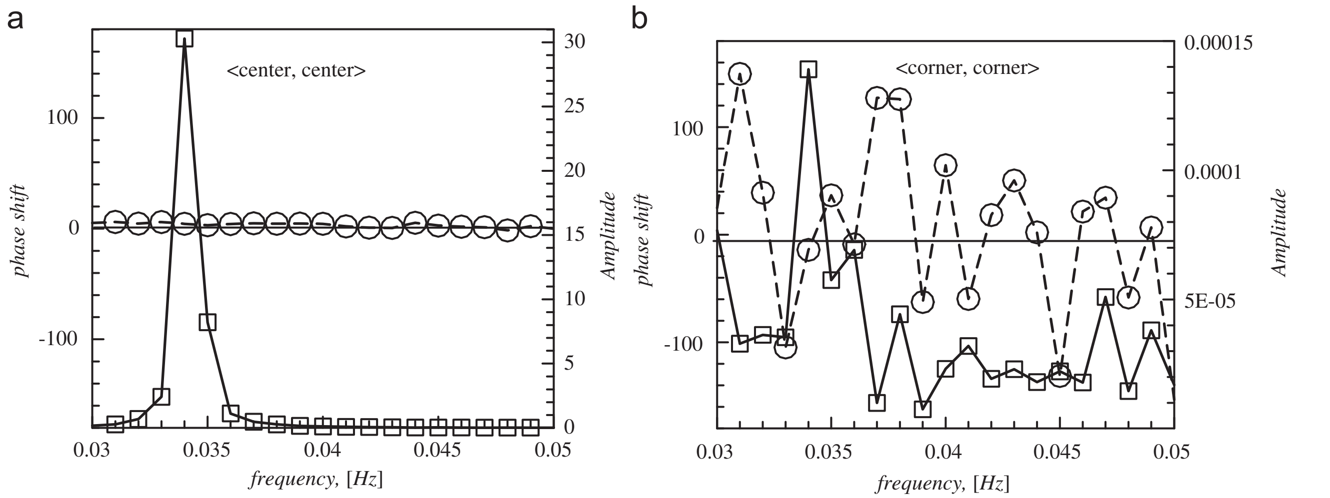

Fig. 6. Amplitude (squares and solid line) and phase shift (circles and dash line) of cross-correlations of the temperature signals obtained by different thermocouples at $\Delta T = 1$ °C. (a) Two thermocouples located in the central part and (b) two thermocouples located in the corner.

studies [5,11]. Also, comparing the frequency values for the stationary and rotating dummy we observe a significant decrease in the oscillation frequency. It can happen because there exist several leading eigenmodes with close growth rates, so that depending on the rotation rate different eigenmodes trigger the instability.

The phase shift between the temperature oscillations at different thermocouples can be used for determination of the developing Fourier mode. In particular, zero phase shift at the maximum correlation amplitude, as appear in Fig. 6, indicate that the axisymmetric mode ($m = 0$) is most probable in this case. Together with the values of critical temperature difference and the oscillations frequency this yields the benchmark data for comparison with numerical predictions. Note, that large-amplitude oscillations measured by thermocouples placed near the container center (Fig. 6a) show zero phase shifts at all frequencies, thus indicating that supercritical flow remains axisymmetric. More surprising are results obtained from the second pair of thermocouples (Fig. 6b). Here, in spite of very small oscillations amplitude, we still succeed to established zero phase shift at the main harmonics $\omega \approx 0.033$ Hz.

Results of computations of critical temperature difference and the corresponding oscillations frequency are shown in Table 2 for azimuthal wavenumber m varying from 0 to 10 for the case without crystal rotation. The computations were done on the grid having 100 and 300 nodes in the radial and axial directions, respectively. It is seen that the calculated critical temperature difference is an order of magnitude larger than that observed in the experiments. The reasons for that are yet to be studied and may be connected, for example, with the surface contamination or with a deep subcriticality of the flow. At the same time, numerically predicted oscillations frequency agrees considerably well with the experimentally measured one. Both experiment and computation predict the same critical Fourier mode, $m = 1$.

Table 2
Numerical results on the instability onset.

m	ΔT_{cr} (°C)	f_{cr} (Hz)
0	7.65	0.0307
1	6.36	0.0263
2	7.67	0.0303
3	7.90	0.0299
4	8.07	0.0294
5	8.25	0.0291
6	8.44	0.0285
7	8.63	0.0277
8	8.84	0.0269
9	9.08	0.0259
10	9.39	0.0264

4. Concluding remarks and discussion

We have developed a new experimental setup that allows us to measure oscillatory instability threshold in a model of Czochralski melt flow. The experiment is aimed to yield data for validation of numerical codes, so that special care is taken to make all the boundary conditions numerically reproducible.

The measured critical temperature difference, oscillation frequency, and number of the most unstable Fourier mode are well-defined values that can be compared with numerical results. All three numbers strongly depend on the flow pattern. Basing on [9,10] we assume that if all three numbers agree the numerical code can be considered as validated at least at the values of ΔT close to critical. For further code validation, comparisons of the whole frequency spectrum can be considered. It is stressed again that if all three parameters are validated, all other flow properties can then be extracted from the validated numerical results.

At the current stage of our research the experimental and numerical results exhibit certain qualitative agreement. The latter can be stated on the basis of the following: (i) strong temperature oscillations were measured at numerically predicted locations; thermocouples placed at locations where numerics predicts very weak oscillations did not register any time-dependent signal; (ii) the critical temperature difference significantly reduces under effect of a weak rotation, as is predicted by our previous studies [11]; (iii) experimental and numerical oscillations frequency and critical Fourier mode are in a good agreement. We assume that we observe the same instability pattern, but destabilized additionally either by a strong surface contamination of water, or by another experimental imperfection. Apparently, programming bugs and computational inaccuracies never can be ruled out. To confirm or to rule out the possibility of surface contamination we plan to perform experiments with other liquids (e.g., silicon oils) or even with an additional no-slip ring replacing the free surface.

Acknowledgements

This study was supported by the German–Israeli Foundation, Grant no. I-954-34.10/2007 and by the Israel Science Foundation, Grant no. 964/05.

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