Crystallization of gallium in a traveling magnetic field as a model experiment

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Abstract
This contribution focuses on small-scale model experiments for directional solidification processes using a gallium volume with a square horizontal cross-section and dimensions of 10x10x7.5 cm^3. A vertical temperature gradient is generated using a heater at the top and a cooler at the bottom. The setup is placed in a coil system generating a traveling magnetic field. The position and shape of phase interface and the melt flow during melting and solidification processes are investigated both experimentally and numerically. A coupled 3D numerical model of melt flow and phase interface is validated.

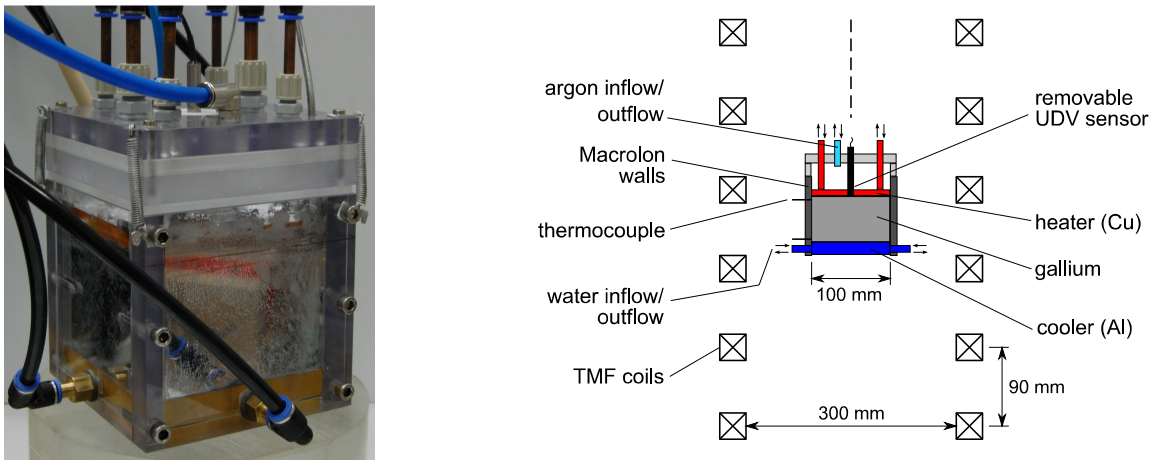
Key words: crystal growth; directional solidification; magnetic fields; model experiments; gallium

Introduction

The directional solidification process is used in the photovoltaic industry to produce large square silicon ingots with a weight up to 1000 kg. This process is performed in a crystallization furnace containing a square-shaped silica crucible surrounded by several heaters at the side or top and a heat sink at the bottom. Silicon raw material, typically in form of chunks, is placed in the crucible and melted. Then the heating power is adjusted to ensure directional solidification of the silicon melt starting from the crucible bottom. The solidification process has a strong impact on the ingot quality and is characterized by high physical complexity [1−2]. The alternating currents in the heaters usually generate Lorentz forces in the molten silicon which interact with buoyancy forces and cause an intense convection. The deformed temperature field in the melt results in a deflected phase interface which may have a significant influence on the flow pattern itself. This interaction process is continuously changing with the melt height which gradually decreases to zero. Numerical simulation is an efficient tool for the understanding and optimization of such complex phenomena especially if it is supported by model experiments to validate the numerical models. This contribution presents such small-scale model experiments using gallium as a model material and a traveling magnetic field (TMF). Pure gallium has a melting point of 29.8 °C, allows for an induction of Lorentz forces similarly to a silicon melt, and can be also solidified in a controlled manner [3−5].

Experimental setup

Model experiments for a TMF-induced flow in a square-based melt container have been performed previously using GaInSn both in an isothermal setup [6] and with a vertical temperature gradient [7]. The present experimental setup (see Fig. 1) is particularly developed for the directional solidification of gallium. The sidewalls of the container are made of



(a) (b)

Fig. 1: Photo of the experimental setup without TMF coils (a); sketch of the setup (b).

Macrolon with a thickness of 15 mm bolted with several screws in the corners. A cooler of aluminum serves as a bottom of the container. It has a thickness of 23 mm and contains a cylindrical cavity with a diameter of 80 mm, located 5 mm from the top surface and 10 mm from the bottom surface. Two water inlets are located at two opposite sides and two outlets at the other two sides. The heater on the top of the melt consists of two rectangular copper tanks glued together. The total thickness is 11 mm with walls of 5 mm on the melt side and 2 mm on the opposite side. Four inlets are located in the corners and two outlets at side centers. The heater and the cooler are connected to *LAUDA* thermostats with typical water flow rates of 3.3 l/min and 2.3 l/min and typical powers of 3.5 kW (heating) and 1.5 kW (heating)/ 0.18 kW (cooling), respectively. The surfaces of heater and cooler are electrically insulated using a *Thermiflex* foil of 0.07 mm thickness. There is a closed volume above the heater flushed with fresh argon to reduce the oxidation of gallium.

Temperature in gallium is measured through the sidewalls with two *Type K* thermocouples located 2 mm from the heater in the center and 2 mm from the cooler at the side wall. A commercial UDV measurement system DOP2000 from *Signal Processing SA* with an 8 MHz probe is used for measurements of the velocity profile and the position of the phase interface through a central opening in the heater. This opening is also occasionally used to mechanically measure the interface position. The phase interface shape is obtained from crystal height measurements at several positions after stopping the experiment and removing the heater (and in some cases also the melt).

Numerical model

The actual numerical model for 3D simulations has been already published in Refs. [1−2]. The 3D time-averaged Lorentz force induced by the TMF in the melt is calculated using the finite element package *GetDP* by solving the time-harmonic equations for the electric and magnetic potentials V and A. The electromagnetic model for the experimental setup was validated using magnetic field measurements. A geometric approximation was developed for the heater and cooler in form of massive plates with effective thicknesses of 4 mm and 8 mm, respectively, with a 2 mm gap to gallium. Electrical conductivities of $3.9 \cdot 10^6$ S/m for liquid $4.7 \cdot 10^6$ S/m for solid gallium (exhibiting a distinct anisotropy) were obtained. Other material properties have been summarized in Ref. [2]. The Lorentz force distribution is calculated for a fixed melt height of 38 mm and deformed together with the mesh in solidification calculations.

Coupled unsteady 3D calculations of the melt flow and phase interface are performed using the finite volume package *OpenFOAM*. The equations for incompressible flow (velocity u, pressure p) considering Lorentz forces F_L and buoyancy forces (gravity g) in Boussinesq approximation as well as the equation for the temperature T are solved in the melt domain:

$$\rho\left[\frac{\partial \vec{u}}{\partial t} + (\vec{u}\cdot\nabla)\vec{u}\right] = -\nabla p + \eta\Delta\vec{u} + \vec{F}_L - \beta(T - T_0)\rho\vec{g}\,, \qquad \nabla\cdot\vec{u} = 0\,, \qquad \rho c\left[\frac{\partial T}{\partial t} + (\vec{u}\cdot\nabla)T\right] = \lambda\Delta T\,,$$

where ρ, η, β, λ, c denote density, viscosity, thermal expansion coefficient, thermal conductivity, and heat capacity respectively. The heat conduction equation is solved in the crystal domain. Both domains are coupled with an isothermal phase interface which is moved/ deformed in each time step according to the local heat balance (Stefan condition) [1]. No turbulence model is used in flow calculation, the Reynolds number in the present setup is approximately Re=1400. The side walls are assumed adiabatic. The heater and cooler surfaces are described according to $\lambda\,\partial T/\partial n = p\,(T - T_W)$ with p the heat transfer coefficient and T_W the thermostat temperature. Values of p=850 W/m^{2}K for the heater and p=1400 W/m^{2}K for the cooler were obtained from stationary temperature measurements [2].

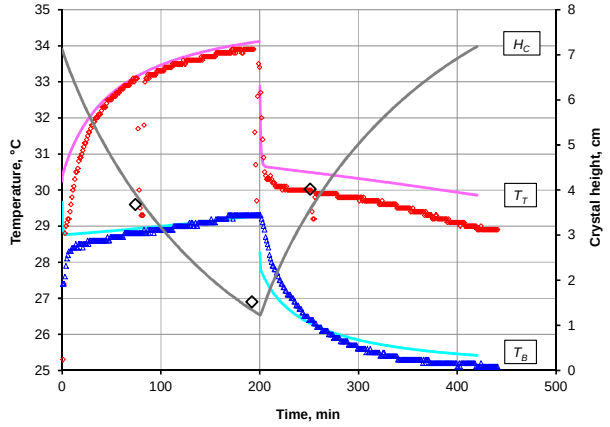
Results and discussion

A typical experiment in the present study starts with a completely solidified gallium. The heater and cooler thermostats are set to T_H =36.9 °C and T_C =28.3 °C, respectively, which initiates a melting process at the top. After the remaining crystal height in the center reaches about 1 cm, the temperatures are switched to T_H =31.2 °C and T_C =23.6 °C, so that a solidification process starts. Fig. 2(a) summarizes the temperatures at the top and bottom of gallium as well as the phase interface position in the center for the case without a TMF (without melt flow). It can be seen that melting and solidification times for 6 cm of crystal height are both about 200 min, with a good agreement between experiment and simulation. Some deviations can be observed for the temperature curves:

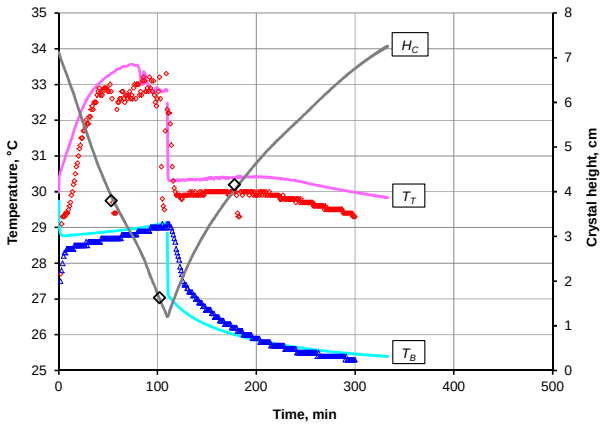
- The simulation was started from a homogeneous temperature distribution in gallium while a small temperature gradient was applied in the experiment. This deviation has no significant influence on the following process.
- The drop of the bottom temperature after switching to solidification is less steep in the experiment. This effect is caused by the limited cooler power and could be included in the simulation with a time-dependent T_C value.
- The top temperature is about 0.5 °C lower in the experiment. This deviation probably occurs due to the melt undercooling which was neglected in the simulation.

Measurements of the interface shape for the case without TMF indicated deviations from a flat surface up to about 3 mm, which is probably caused by the anisotropic thermal properties of solid gallium.

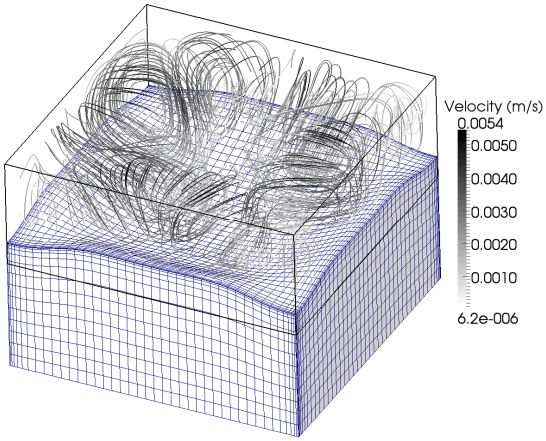
The previous experiment was repeated with a TMF up using a frequency of 16.6 Hz, phase shift of 60°, and current amplitude of 705 A in each coil. The resulting Lorentz force distribution is shown in Fig. 2(d) and reaches a maximum force density of 40 N/m^3. The 3D streamlines for a middle melt height in Fig. 2(c) generally indicate a toroidal flow



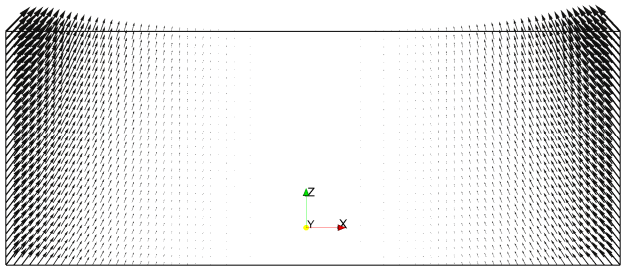
(a) No TMF



(b) TMF up

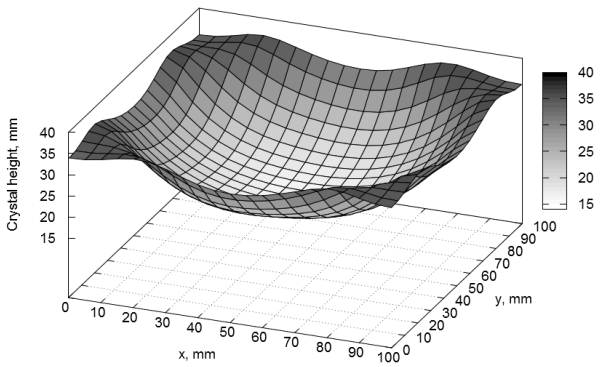


(c) TMF up

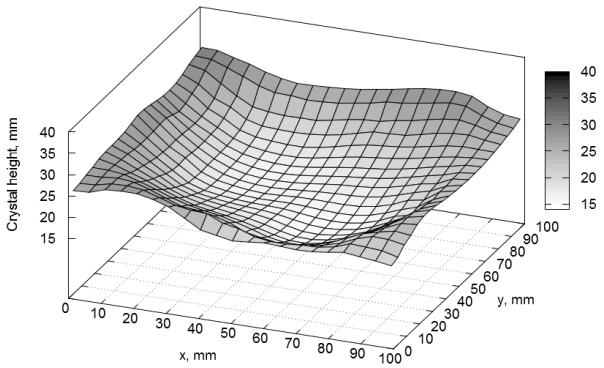


(d) TMF up, $F_{L\,MAX}$ (cut) = 39.7 N/m^3

Fig. 2: Experiments and simulations for Ga melting/ solidification without TMF and with TMF up: top (T_T) and bottom (T_B) temperatures, crystal height (H_C) in the center (a,b) where experimental results are shown with symbols ◊; 3D flow streamlines (c) and Lorentz force distribution in vertical cross-section (d) for melt height H_M=38 mm.

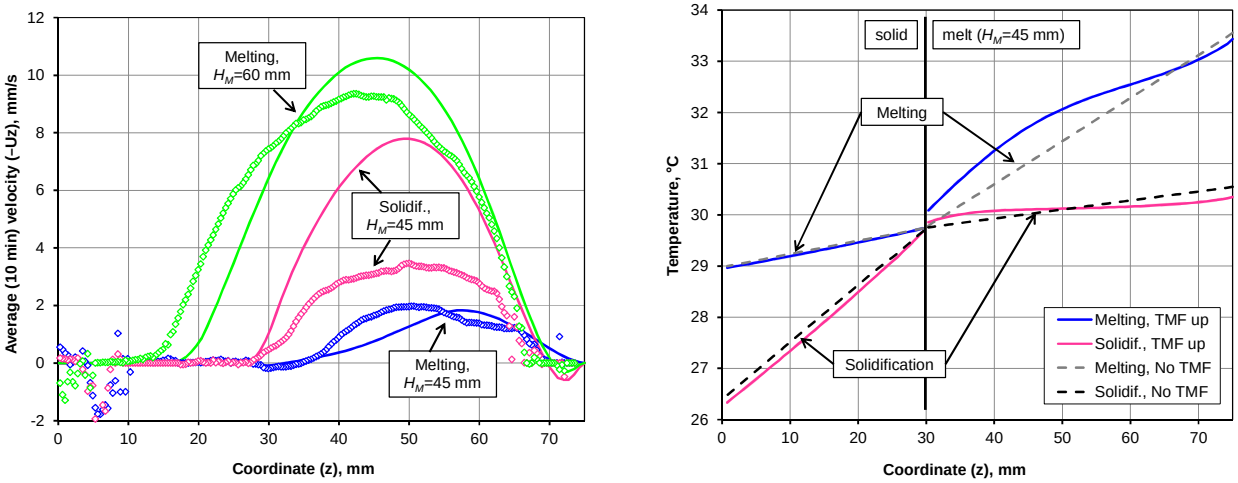


(a) Simulation



(b) Experiment

Fig. 3: 3D interface shape with TMF up at the end of melting with a remaining crystal height of 15 mm in the center: numerical results (a) and experimental measurements at 15x15 points (b).



(a) (b)

Fig. 4: Ga melting/ solidification: time-average velocity with TMF up (a) and temperature profiles without TMF and with TMF up (b) on the vertical axis (z=0 on crystal bottom) for various melt heights H_M. Experimental results are shown with symbols ◊.

structure already known from similar experiments [6−7], but in this case the torus is broken in the corners. The vertical velocity profiles in Fig. 4(a) confirm a downward flow in the center. Note that reproducible velocity measurements at the central position were possible only during solidification and at large melt heights, when the velocity magnitude reached about 10 mm/s. Since the vertical temperature difference in the melt was about 0.3 K during solidification and 3.5 K during melting (see Fig. 4(b)), damping of the TMF flow by the buoyancy forces was much stronger during the melting stage.

The time-dependence of interface position in Fig. 2(b) shows that the melting time in the center is reduced due to the TMF flow to about 100 min while the solidification time remains approximately the same. The time-dependence and distribution of the temperature in gallium is qualitatively similar to the case without a TMF, see also Fig. 4(b). The most distinct effect of a TMF flow can be observed in the changes of phase interface shape. Fig. 3 demonstrates that at the end of melting the interface becomes concave, with a maximum deflection of 15 mm. The four corners exhibit a local convex deflection, and the whole interface shows a slight asymmetry in the experiment. This asymmetry is probably caused by the anisotropic crystal properties. Apart from that, the measurements agree well with the calculated interface shape. Note that the melt flow velocity during the melting was relatively weak reaching only about 2 mm/s in the central part, see Fig. 4(a).

Conclusions

It can be concluded that already relatively weak melt flows induced by time-dependent magnetic fields such as a TMF may lead to significant changes of the local melting rate and consequently a deformation of the phase interface. Such effects should be taken into account for silicon growth processes and can be efficiently investigated using unsteady 3D flow simulations validated in small-scale model experiments. A good agreement between experimental and numerical results was achieved in the present work.

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