ELSEVIER

Physica A 249 (1998) 190–195

PHYSICA A

Interferometric measurement of the temperature field in the vicinity of ice crystals growing from supercooled water

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Abstract

Ice crystals are grown in the supercooling temperature range of 0–8°C, where the crystal-growth morphology shows a dependence on temperature. In order to understand the growth mechanism, we measure the temperature field around the growing crystals by using the temperature dependence of the refractive index. Since this has a zero for H_2O at 0°C, we use D_2O, which has similar growth morphologies, and achieve considerably greater sensitivity. The crystal growth cell lies within four imaging Mach–Zehnder interferometers, which observe it in different directions. From the interferograms the three-dimensional temperature field is deduced. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

The growth of ice crystals from supercooled water is a classic example of growth of a highly anisotropic crystal from its melt. The morphologies of these crystals have been studied in detail [1–6]. Two questions which particularly interest us concern the characteristics of a two-dimensional crystal growing in a three-dimensional medium, which has received little theoretical attention, and the origin of pyramid pairs (Fig. 1) which grow at supercooling > 2.7°C, discussed by Ryan [5,6] in 1966 who gave only a very qualitative explanation.

In order to gain a deeper understanding of the kinetics of the growth of crystal morphologies, it would be an advantage to have a picture not only of the crystal shape itself, but also of the fields driving the growth. In the present case, the driving field is a temperature field $T(r)$, which can be generally described as having a value of $T_m - \Delta$ at infinite distance, rising at the crystal surface to a value $T_m - \delta$ close to, but still below, the melting temperature T_m [7,8]. The driving force for crystal growth is

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PII S0378-4371(97)00465-2

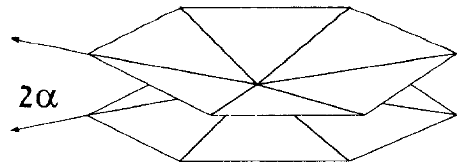

Fig. 1. Double pyramidal growth of ice crystal.

the local-surface supercooling δ, and the latent heat is extracted by thermal diffusion through the water; these two processes control the crystal morphology. In the parallel case of crystal growth from solution, where the concentration of the solution controls the crystal growth, optical interference methods have been used to measure the concentration field, usually under conditions where the temperature field can be neglected. This was done by the use of a thin cell with isothermal boundaries. Such experiments, carried out under an interference microscope, were first done by Goldstaub [9] and the methods have recently been made quantitative [10–13]. Since the two-dimensional microscope field of view represents integration through a three-dimensional concentration field, models of the field were used in order to relate the observations to the actual field around the crystal, such as an assumption of cylindrical symmetry [13].

When considering growth of a pure crystal from its melt, the problem is intrinsically simpler, and much headway has been made using plastic solids such as succinonitrile [14]. However, the controlling field, in this case temperature, has never been measured. Clearly, such experiments cannot be performed in a thin cell but must be conducted in an essentially infinite three-dimensional medium. The temperature at each point can be sensed by its effect on the refractive index, but optically, one can only measure variations of refractive index integrated along a complete optical thickness. To measure local values of the three-dimensional refractive-index field (i.e. temperature field) one should observe it simultaneously from several directions and use some form of tomographic reconstruction [15].

2. Refractive index of water

The key to measurement of the temperature field is the dependence of refractive index of water on temperature. On the one hand, if the dependence is weak, it is difficult to make measurements with sufficiently high signal-to-noise ratio in order to carry out a tomographic analysis. On the other hand, if the dependence is too strong, or the temperature gradient too high, a light ray through the experimental region is bent and the consequent geometrical problem becomes very difficult. Water is peculiar in that its refractive index is not a linear function of temperature in the region of 0°C. One might expect from the Clausius–Mossotti relation that the refractive index is maximum at 4°C (as is the density) and varies approximately parabolically about this temperature. This is not so; the maximum refractive index of water occurs close to 0°C

[16], a fact which is not well understood. This is unfortunate from the point of view of the present experiment for two reasons: first because it makes the gradient dn/dT very small in the region of $T = -4^\circ C$ to $0^\circ C$, and second because the dependence in this region cannot be approximated as linear, which complicates the data analysis. Fortunately, for the experiments, a change from regular water to heavy water, D_2O, solved the above problems. Heavy water has its maximum density at 11.2°C, maximum refractive index at 6.75°C and melting point at 3.82°C. We confirmed that the growth morphologies occurring in regular ice also occur in heavy ice.

The refractive index [17] in the region of interest can be expressed for D_2O by

$$n(T) = n_0 + n_1(T - T_{MI})^2 + n_2(T - T_{MI})^3 ,$$

where T_{MI} is the temperature of maximum refractive index, 6.75°C, $n_0 = 1.329262$, $n_1 = -2.77\,10^{-6}$, $n_2 = 1.68\,10^{-8}$.

3. Optical tomography

The use of tomographic reconstruction from optical interferometric data has been intensively investigated in recent years [18,19]. It is desirable to obtain data in which the optical path is integrated along as many different directions as possible, and this has been achieved in several experiments by using holographic interferometric reconstruction. However, ice crystals grow quickly, and we were faced with the need to record a sequence of interferograms in different directions in quick succession, for which purpose holography is inappropriate. For this purpose, the experimental cell is sampled by four Mach–Zehnder interferometers which image the cell from directions at 45° to each other in the horizontal plane. The interference patterns are recorded simultaneously by four video cameras and phase shifts are extracted by Fourier fringe analysis [13]. Four is the smallest number of views which has been used successfully for tomographic reconstruction [19]. We use an algebraic reconstruction technique [20] (ART) developed by Verhoeven [18]. In some regions of the experimental cell one or more of the views is obscured by the growing crystal. The tomographic part of this work is still under development, but an accuracy of about 0.1 K in a 1 mm^3 volume has been achieved.

4. Nucleation of crystals

It is not trivial to nucleate ice crystals from very pure water at small degrees of supercooling. Below two degrees nucleation is achieved by electrofreezing [21,22], which uses a high-voltage electric field pulse between sharp electrodes to nucleate the solid. At smaller degrees of supercooling, we have developed a nucleation device which relies on both electrofreezing and a temperature gradient [23]. When the ice is nucleated within a capillary tube, a single-crystal emerges from its tip.

5. Results and discussion

The shape of an ice crystal at small supercooling (<2.7°C) is quasi-two dimensional i.e., a flat crystal, with a rich in-plane structure having hexagonal symmetry. The envelope of the crystals is shape preserving [7]. The two-dimensional ice crystals grow in a three-dimensional medium and the temperature field which is developed in this situation has a characteristic form. Fig. 2 compares a simulation [1] with experimentally determined projection of the temperature field.

At higher supercooling (>2.7°C) the ice grows in a different morphology. Instead of six $\langle 1\,1\,\bar{2}\,0\rangle$ growth directions in the plane normal to the hexagonal axis, a 12 sided double pyramid (dodecaconal) is observed with growth along non-rational directions $\langle 1\,1\,\bar{2}\,z\rangle$ and $\langle 1\,1\,\overline{2z}\rangle$, where z increases with the supercooling and has typical value 0.2 at >5°C supercooling. Fig. 3 shows the temperature field as measured between the two pyramids compared with an appropriate simulation. Brener and Levine [24] suggested on theoretical grounds that anisotropic crystals could select low-symmetry growth directions if the orientations of minimum surface tension and minimum kinetic growth coefficient were not related by reflexion symmetry. They give a two-dimensional example, which has fourfold symmetry, and the two minima are at arbitrary orientations to one another (not 90° or 45°). They show that the selected growth orientation then depends continuously on the degree of supercooling. We suggest that the double-pyramidal growth of ice crystals may be an example of such behaviour, and measurement of parameters of this type of growth will allow comparison with their model.

Convection in water can be observed as an asymmetry in the temperature field (Fig. 4). It should be recalled that in supercooled water the hotter fluid descends. The influence of convection on the growth shape has also been observed.

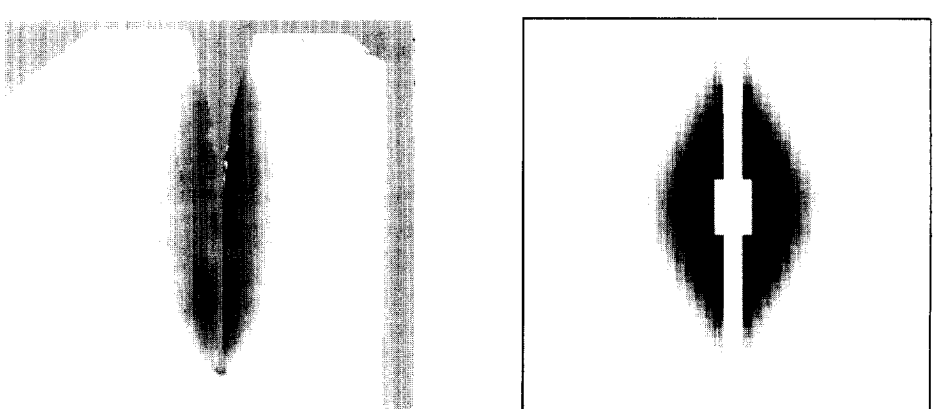

Fig. 2. Temperature field around plate crystal; experiment and simulation.

[1] Simulations were carried out for a two-dimensional crystal with square symmetry, not hexagonal. Heat is released from the growing edges into a three-dimensional medium. They are for qualitative comparison only.

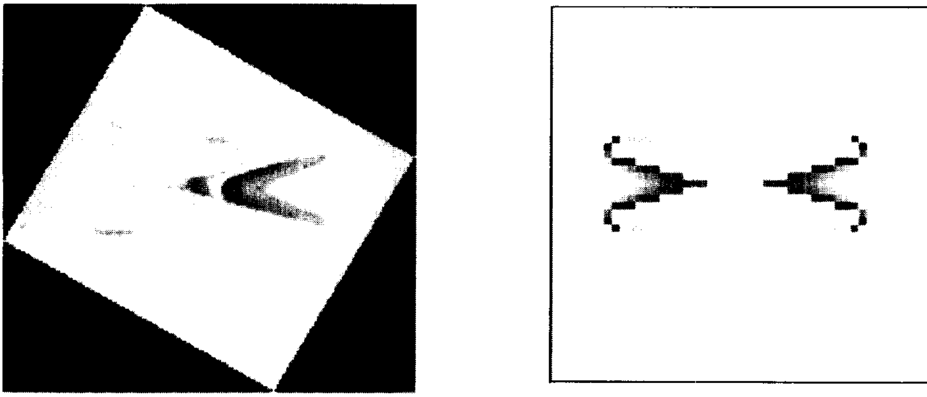

Fig. 3. Temperature field between two pyramids; experiment and simulation.

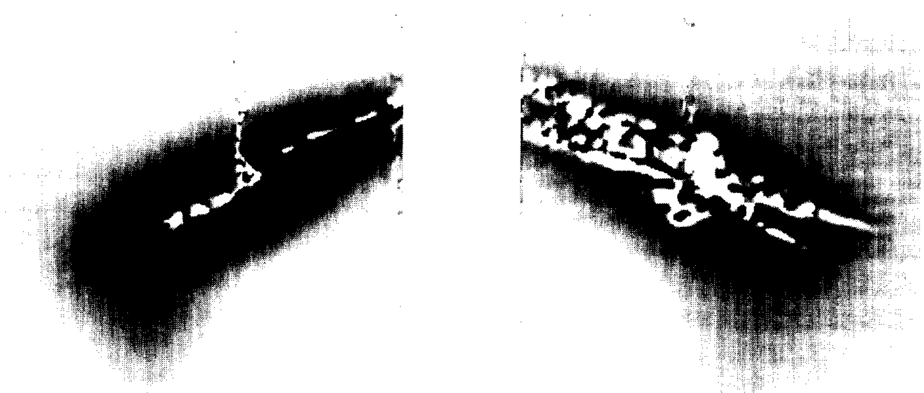

Fig. 4. Convection in the temperature field.

6. Summary

These experiments represent the first direct observation of temperature fields around growing crystals. There still remain considerable difficulties in deriving the complete three-dimensional fields by tomography because of the small size of the observed signals. Some features of the field can be related to the integrated field, without need for reconstruction.

Acknowledgements

We acknowledge helpful discussions with Raz Kupferman and Dean Verhoeven, who also supplied us with algorithms for the tomographic reconstruction. This research was partially supported by the German–Israel Binational Science Foundation (GIF), the

Fund for Basic Research of the Israel Academy of Sciences and the Minerva Center for Nonlinear Research.

References

[1] K.K. Koo, R. Ananth, W.N. Gill, Phys. Rev. A 44 (1991) 3782.
[2] P.V. Hobbs, Ice Physics, Clarendon Press, Oxford, 1974.
[3] J.S. Langer, R.F. Sekerka, T. Fujioka, J. Crystal Growth 44 (1978) 414.
[4] Y. Furukawa, W. Shimada, in: N. Maeno, T. Hondoh (Eds.), Physics and Chemistry of Ice, Hokkaido University Press, Sapporo, 1992.
[5] W.C. Macklin, B.F. Ryan, Philos. Mag. 14 (1966) 847.
[6] W.C. Macklin, B.F. Ryan, J. Atmos. Sci. 22 (1965) 452.
[7] R. Kupferman, Morphology, coexistence and selection in interfacial pattern formation, Ph.D. Thesis, Tel-Aviv University, 1995.
[8] E. Brener, H. Müller-Krumbhaar, D. Temkin, Phys. Rev. E 54 (1996) 2714.
[9] F. Mussard, S. Goldsztaub, J. Crystal Growth 14 (1972) 445.
[10] E. Raz, S.G. Lipson, E. Polturak, Phys. Rev. A 40 (1989) 1089.
[11] A. Tanaka, M. Sano, J. Crystal Growth 125 (1992) 59.
[12] Ph. Bouissou, A. Chiffaudel, B. Perrin, P. Tabeling, Europhys. Lett. 13 (1990) 89.
[13] S. Kostianovski, S.G. Lipson, E.N. Ribak, Appl. Opt. 32 (1993) 4744.
[14] S.C. Huang, M.E. Glicksman, Acta Metall. 29 (1981) 701.
[15] D.W. Sweeney, C.M. Vest, Appl. Opt. 12 (1973) 2649.
[16] Ch. Saubade, J. Phys. 42 (1981) 359.
[17] H. Eisenberg, J. Chem. Phys. 43 (1965) 3887.
[18] D. Verhoeven, Appl. Opt. 32 (1993) 3736.
[19] D. Mewes, R. Renz, Chem. Ing. Technol. 63 (1991) 699.
[20] G.T. Herman, A. Lent, S.W. Rowland, J. Theor. Biol. 42 (1973).
[21] H.R. Pruppacher, Pure Appl. Geophys. 104 (1973) 623.
[22] I. Svishchev, P.G. Kusalik, J. Am Chem. Soc. 118 (1996) 649.
[23] I. Braslavsky, S.G.Lipson, submitted to Appl. Phys. Lett.
[24] E. Brener, H. Levine, Phys. Rev. A 43 (1991) 883.