Formation of brine channels in sea ice

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Abstract. Liquid salty micro-channels (brine) between growing ice platelets in sea ice are an important habitat for CO2-binding microalgaea with great impact on polar ecosystems. The structure formation of ice platelets is microscopically described and a phase field model is developed. The pattern formation during solidification of the two-dimensional interstitial liquid is considered by two coupled order parameters, the tetrahedricity as structure of ice and the salinity. The coupling and time evolution of these order parameters are described by a consistent set of three model parameters. They determine the velocity of the freezing process and the structure formation, the phase diagram, the super-cooling and super-heating region, and the specific heat. The model is used to calculate the short-time frozen micro-structures. The obtained morphological structure is compared with the vertical brine pore space obtained from X-ray computed tomography.

1 Introduction

Sea ice does not freeze homogeneously but some liquid salty micro-channels remain which are called brine. These brine capillaries are an important habitat for CO2-binding microalgae with great impact on the polar ecosystems. Their carbon consumption amounts to about 18% of the entire carbon consumption in the southern ocean. Therefore it is desirable to understand the formation of such brine channels as possible habitat for carbon-binding algae. Two-phase regions of pure ice crystals and water are also known as mushy layers in the context of binary alloys [1, 2]. Highest cell abundances occur in these regions, due to the higher porosity and due to the constant flushing with nutrient-rich seawater [3, 4].

The freezing process of salty water is one example of the solidification of binary alloys [5, 6]. Models of ice polluted with any salt as “liquid jelly” [7] consider this process as first-order phase transitions [8]. Sometimes, for solidification of seawater, the model of percolation transitions is used in brine trapping [9–11]. In this respect a morphological stability analysis was applied to the solidification of salty water [12]. All these quantitative models [13–15] have investigated the brine channel volume, salinity profile or heat expansion, but have unfortunately not considered the pattern formation. Here we will present a dynamical model exploring the formation of morphological patterns consistent with the thermodynamics of freezing. Concentrating on the short-time evolution we consider the structure-forming processes here as adiabatic and neglect the heat transport.

Images of single crystals in sea ice with the help of X-ray computed tomography [16] show arrays of nearly parallel brine layers whose connectivity and complex morphology vary with temperature. The pore space turns out to be much more complicated than suggested by simple models of parallel ice lamellae and parallel brine sheets [17]. Sometimes the granular sea ice texture is imagined to arise from a deposition of fragile ice crystals. They are thought to be formed within the turbulent ocean interior and then rising buoyantly to the ocean surface [18, 19]. In these settings the size of the settling crystals plays a dominant role in controlling the observed structures. We consider here the opposite view that these structures result from a thermodynamic instability during growth itself rather than from the external deposition.

In order to describe a realistic pattern formation and the phase transition on the same theoretical basis we use a phase-field model for the solidification of the two-dimensional interstitial liquid. We will calculate the frozen micro-structures and will compare with the vertical brine pore space obtained from X-ray computed tomography [16, 20]. The aim is to present a model with the smallest possible number of microscopic parameters to be extracted from experiments. We find here that three parameters are sufficient, the freezing, the structure, and

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the diffusivity parameter. Only the first two determine the phase diagram while the diffusivity enters the brine channel size. The linear stability analysis leads then to the parameter range where structure can appear and the numerical solution will allow to compare with the experimental data.

The outline of the paper is as follows. First we develop the minimal model and give the meaning of different used model parameters. Then we derive the thermodynamics of supercooling and freezing point depression providing the phase diagram. In sect. 4 we will discuss the linear stability analysis which yields the most unstable modes and scales. Then we determine the model parameters from the properties of water in sect. 5. The time evolution is presented by a numerical solution of the coupled phase-field model in sect. 6 and is compared with the experiment in sect. 7. Section 8 summarizes and discusses shortcomings as suggestions for further investigations.

2 Phase-field model

To distinguish between ice and water molecules we use a two-state function, the “tetrahedricity” [21]

$$u = 1 - \frac{1}{15(l^2)} \sum_{i,j} (l_i - l_j)^2,$$

(1)

where the $l_i$’s are the differences of the six edges of the tetrahedron formed by the four nearest neighbors of the considered water molecule. For an ideal tetrahedron one has $u = 1$ and the random structure is represented by $u = 0$. We assume the standard expansion of the energy function in powers of this order parameter [8, 22]

$$\frac{D_{\text{ice}}}{2} (\nabla u)^2 + \frac{a_1}{2} u^2 - \frac{a_2}{3} u^3 + \frac{a_3}{4} u^4 + h \frac{2}{\alpha} v^2 + D_{\text{salt}} \frac{2}{2} v^2.$$

(2)

Here we have coupled additionally a second-order parameter, the salinity $v$, by the term $h$ which can be considered as reaction rate between water and ice. The parameter $a_1$ is the freezing parameter determining the phase transition, the structure parameter $a_3$ is responsible for the non-linear behavior and $D_{\text{ice}}$ and $D_{\text{salt}}$ are the diffusion coefficients of ice and salt. The coefficient $a_2$ is connected with an uneven exponent and is therefore responsible for the phase transition of first kind. All these parameters depend on the temperature and can be scaled to only three relevant parameters. The phase diagram will be determined only by two of them, the dimensionless structure and the freezing parameter.

The coupling of the two order parameters is chosen in a form which enables the conservation of the total mass of the salt as follows. We demand a balance equation of the form $\partial \psi / \partial t = -\nabla \psi$ where the current is assumed to be proportional to a generalized force $\psi \sim F$ which should be given in terms of a potential $F = -\nabla \psi$. This potential in turn is expressed by the variation of the free energy density $P = \delta f / \delta v$. This procedure is nothing but the second law of Fick and we obtain an equation of the Cahn-Hilliard-type without the fourth derivation for the evolution of the salinity $v$.

Defining the reduced time $\tau = D_{\text{salt}} a_2^2 / h^2$, the spatial coordinates $\xi = a_2 x / h$, the dimensionless order parameters of ice/water structure $\psi = h^3 \rho / D_{\text{salt}} a_2$, and the salinity $\rho = h^3 v / D_{\text{salt}} a_2$, we obtain the coupled order parameter equations

$$\frac{\partial \psi}{\partial \tau} = -\alpha_1 \psi + \psi^2 - \alpha_3 \psi^3 - \psi \rho + D \frac{\partial^2 \psi}{\partial \xi^2},$$

$$\frac{\partial \rho}{\partial \tau} = \frac{1}{2} \frac{\partial^2 \psi}{\partial \xi^2} + \frac{\partial^2 \rho}{\partial \xi^2}.$$  

(3)

These time-dependent Ginzburg-Landau differential equations couple the dynamics of the dimensionless order parameter $\psi$ and the dimensionless salinity $\rho$ depending only on three parameters, the freezing parameter $\alpha_1 = a_1 h^2 / a_2^2 D_{\text{salt}}$, the structure parameter $\alpha_3 = a_3 D_{\text{salt}} / h^2$, and the diffusivity $D = D_{\text{ice}} / D_{\text{salt}}$ with $\alpha_1$, $\alpha_3$, $D > 0$. Equations (3) represent a modification of the model C in the Hohenberg-Halperin classification [23], there eq. (4.50). The difference here is an additional quadratic term in the first equation coming from the uneven exponent in (2) with $a_2$ responsible for the first-order phase transition. We neglect in this model any velocity or temperature field which could be included analogously to the model H in [23].

3 Thermodynamics of supercooling, super-heating and freezing point suppression

The parameters $\alpha_1$ and $\alpha_3$ describe the regions of ordered and non-ordered phase. This can be seen from the uniform stationary free energy density. We therefore use the stationary solution of the second equation in the first one of (3) to obtain

$$f(\psi_0, \rho_0) = \frac{\alpha_1}{2} \psi_0^2 - \frac{1}{3} \psi_0^3 + \frac{\alpha_3}{4} \psi_0^4,$$

(4)

where the temperature-dependent compound parameters $\alpha_1(T) = \alpha_1(T) + \rho_0$ and $\alpha_3 = \frac{1}{2} \rho_0^2 - \gamma \rho_0$ appear in terms of the salinity $\rho_0$. Freezing-point depression occurs since $\alpha_1' + \rho_0$ corresponds to a higher temperature than $\alpha_1$.

The temperature and salinity dependence of $\alpha_3$ is supposed to be weak near the phase transition. At the lower limit of the super-cooling region of fresh water [24, 25], $T_0 = 233.15 K$, the parameters $\alpha_1$ vanishes linearly for first-order phase transitions [22] such that we can assume $\alpha_1(T) = \alpha_1(T - T_0)$. The freezing point depression in the framework of Landau-Ginzburg theory can be expressed therefore as

$$\Delta T = -\frac{\rho_0}{\alpha_1} = -\frac{D_{\text{salt}} a_2^2}{a_1} \frac{\Delta T}{h^2} \rho_0.$$

(5)

Introducing the salinity-dependent super-cooling temperature $T_{c,s} = T_0 - |\Delta T|$ the freezing parameters $\alpha_1$ depends
on the temperature according to
\[ \alpha_1(T) = \rho_0 \frac{T - T_{c,s}}{|\Delta T|}. \] (6)

The free energy density (4) has a minimum at \( \psi_0^0 = 0 \) and a minimum/maximum for
\[ \psi_0^\pm = \frac{1}{2\alpha_3} \left(1 \pm \sqrt{1 - 4\alpha_1\alpha_3}\right). \] (7)

For \( \alpha_1 > 1/4\alpha_3 \), the minimum at \( \psi_0^0 = 0 \) is the only allowed physical solution, which is the disordered state. As long as
\[ \alpha_1 \leq \frac{1}{4\alpha_3} = \alpha_1(T), \] (8)
a second relative minimum appears at \( \psi_0^+ \) as seen in fig. 1a. The lowest free energy establishes the stable state. The coexistence curve where these two local minima are equal and \( f(\Psi_0^+) = f(\Psi_0^0) = 0 \) yields the critical temperature
\[ \alpha_1(T_c) = \frac{2}{9\alpha_3}. \] (9)

This coexistence curve is plotted as solid line in fig. 1. Above the critical parameters \( \alpha_1(T_c) < \alpha_1(T) < \frac{1}{4\alpha_3} \) the ordered phase \( \psi_0^+ > 0 \) is metastable whereas the non-ordered phase \( \psi_0^0 = 0 \) is stable. For small \( \alpha_1 \leq \alpha_1(T_c) \) the second minimum at \( \psi_0^+ > 0 \) becomes deeper and the ordered phase \( \psi_0^+ \) is the stable one. Therefore the absolute minimum changes discontinuously from \( \psi_0 = 0 \) to \( \psi_0^+ > 0 \) as plotted in fig. 1(b). The jump at \( T_c \) is a measure for the latent heat during the first-order phase transition between water and ice.

We identify the upper borderline of a stable structure formation (9) with the freezing temperature since this is the line where structure, i.e., ice formation, is possible at all. In the same manner the borderline of metastable structure (8) represents the super-heating temperature. The shaded area in fig. 1 describes the super-cooling region between \( T_2 \) and \( T_c^0 \). The latter one is the temperature where \( \alpha_1 = 0 \). Above this area we find the super-heating region for \( T_2 < T < T_c \). From (8) and (9) the relation between the super-cooling temperature \( T_c^0 \), the freezing temperature \( T_c \), and the super-heating temperature \( T_1 \) reads
\[ T_1 = \frac{9}{8} T_c - \frac{1}{8} T_c^0. \] (10)

4 Linear stability analysis

The linear stability analysis for the two local minima around the disordered phase \( \psi_0^0 \) and the ordered phase \( \psi_0^+ \) with \( \rho_0 \propto \exp(\lambda(\kappa)\tau + i\xi) \) leads to the two possible growth rates
\[ \lambda_{1,2} = -\frac{1}{2}[(D + 1)\kappa^2 - \kappa \pm \sqrt{\Delta}], \] (11)
with \( \Delta = [(D - 1)\kappa^2 - \kappa]^2 + 4\kappa^2 \gamma_1^2 > 0 \) and \( \gamma = -\alpha_1 + 2\psi_0 - 3\alpha_3\psi_0^2 \) which takes the value \( \gamma = -\alpha_1 \) for the fixed point \( \psi_0^0 = 0 \) and \( \Psi = \psi_0 - 2\alpha_3\psi_0^2 \) for \( \psi_0^+ \). Time-oscillating structures would appear only if \( \text{Im} \lambda(\kappa) \neq 0 \), i.e., \( \Delta < 0 \), which is not the case in our model.

An unstable fixed point \( \lambda(\kappa) > 0 \) allows any fluctuation with a wave vector \( \kappa \) to grow exponentially in time. For the fixed point representing the disordered phase, \( \psi_0 = 0 \) and \( \rho_0 = \text{const} \),
\[ \lambda_{1,2} = \frac{1}{2}[-(D + 1)\kappa^2 - \alpha_1 \pm [(D - 1)\kappa^2 + \alpha_1]] < 0, \] (12)
and no structure formation occurs in this state which was expected for the disordered phase, of course.

We can only have positive \( \lambda(\kappa) \) if the values of \( \kappa \) are restricted to the region between the zeros of \( \lambda(\kappa) \), which is \( \kappa^2 \in (0, \psi_0^+ [1 - (2\alpha_3 - 1)\psi_0^+]/D). \) Discussing separately the cases \( \alpha_3 > \sqrt{\frac{1}{2}} \) and \( \sqrt{\frac{1}{2}} \leq \alpha_3 \leq 1 \) and recombining the results, we obtain the range for possible structure formation
\[ 2 > \alpha_3 > 1 \Rightarrow \frac{1}{4\alpha_3} \left(1 - \frac{1}{(2\alpha_3 - 1)^2}\right) < \alpha_1 < \frac{2}{9\alpha_3}, \]
\[ 1 > \alpha_3 > 0 \Rightarrow \alpha_1 < \frac{2}{9\alpha_3}, \] (13)
represented in fig. 2 as a phase diagram for the freezing and structure parameters.

The structure parameter \( \alpha_3 \) determines the brine channel formation. A small \( \alpha_3 \) means low temperatures or low salinities and consequently a freezing process with a uniform ice phase for sufficiently large \( \alpha_3 \) and a precipitate of salt. In contrast at higher \( \alpha_3 \) there are higher temperatures or higher salinities inducing a melting with a uniform liquid water phase and dissolved salt. The spatial structures can only appear in the instability region which starts at the maximal point \( \alpha_1 = 1/9 \) at \( \alpha_3 = 2 \). The description of the instability region does not involve a restriction on the diffusivities of salt and water. This is different from the model of [26], which describes structure formation in sea ice in terms of Turing structures.

5 Determination of parameters

Before solving (3) numerically we use (6) to determine the values of \( \alpha_1 \) and \( \alpha_3 \) in terms of water properties.

Using the latent heat of the phase transition from water to ice \( \Delta H = 6 \text{kJ/mol} \) and a dissociation ratio of \( x = (n_{\text{Na}^+} + n_{\text{Cl}^-})/n_{\text{H}_2\text{O}} = 1/50 \), the Clausius-Clapeyron relation yields a freezing point depression of \( \Delta T_{\text{ice}} = -x \frac{\Delta H}{2 \rho_l T_0} = -2 \text{K} \) in agreement with the natural value of \( \Delta T = -1.9 \text{K} \). After a super-heating of more than 5°C, homogeneous nucleation occurs in the metastable state [27]. For fresh water \( (T_c = T_0 = 273.15 \text{K} \) and \( T_{\text{c,s}} = T_0 = 273.15 \text{K} \)) from eq. (10) follows that \( T_1 = 278.11 \text{K} \) (4.96°C) as the upper limit of super-heating in agreement with the experiment [27]. According to (10) and (9) and (8), these super-heating and freezing temperatures are realized by choosing \( \alpha_1 = 0.2 \) and \( \alpha_3 = 0.9 \). The structure parameter \( \alpha_3 = 0.9 \) leads to a freezing point temperature of \( -1.9 \text{°C} \) \( (T_c = 271.25 \text{K}) \) for seawater of salinity 35 g/kg \( (\rho_0 = 0.6 \text{mol NaCl/53 mol H}_2\text{O} = 0.0113) \) and represents therefore a realistic description of super-cooling pure water.

Furthermore, the specific heat \( c \) is dependent on \( \alpha_3 \) as

\[
c = -\frac{T \partial^2 f(\psi_0(T))}{\partial T^2} \bigg|_{T=T_0^0} = \frac{\alpha_3^2 T_0}{2 \alpha_3} \left(1 + \frac{3}{\sqrt{1 + 36 \alpha_3 T_0^0}}\right) = \frac{4}{81 \alpha_3^3 (T_c - T_0^0)^{\frac{3}{2}}}.
\]

We set the energy scale to be the difference of the latent heat of water freezing \( K_E = L(0 \text{°C}) - L(-40 \text{°C}) = 98.1\text{J/g} \) [24]. The resulting specific heat in our theory yields \( c_{\text{spec}} = K_E c = 2.14\text{J/gK} \) which compares well with the experimental value of \( c_{\text{exp}} = 2.1\text{J/gK} \). This shows that the choice of the structure parameter \( \alpha_3 = 0.9 \) is in agreement with the specific heat too.

The parameters \( \alpha_1 \) and \( \alpha_3 \) define the local portion of the free energy in a system with uniform order parameters and salinity. The spatial inhomogeneity of the system is described by the third parameter of the model \( D = D_{\text{ice}}/D_{\text{salt}} \). At the freezing temperature of seawater of \(-1.9 \text{°C}\), the study in [28] predicts \( D_{\text{salt}} = 0.62 \times 10^{-5} \text{cm}^2/\text{s} \). The \( D_{\text{ice}} \) can be linked to the reorientation rate of the \( H_2O \)-molecules and the correlation length which leads with realistic numbers [29,30] to \( D_{\text{ice}} = 0.33 \times 10^{-5} \text{cm}^2/\text{s} \) and finally to a ratio \( D_{\text{ice}}/D_{\text{salt}} = 0.47 \).

6 Time evolution and pattern formation

Now we integrate the equation system (3) numerically in one and two space dimensions by an exponential time differencing scheme of second order (ETD2) [31]. We have a stiff differential equation of the type \( \dot{y} = ry + z(y,t) \), with a linear term \( ry \) and a nonlinear part \( z(y,t) \). The linear equation is solved analytically and the integral over the nonlinear part is approximated by a proper finite differencing scheme.

The evolution of the order parameter \( \psi \) and the salinity \( \rho \) in one and two dimensions is shown in fig. 3. The quantities \( \psi \) and \( \rho \) are complementary in phase. Due to the second equation of (3), the conservation of salinity \( \int dx \rho(t,x) = \text{const} \) is ensured. We can absorb this mean salinity into \( \rho \to \rho - c \) leading to a mere shift in \( \alpha_1' - c + \rho_0 = \alpha_1 \) which means we consider with \( \rho \) the deviations from a mean salinity \( c \) and the total salinity remains positive. Regions of high salinity correspond to the water phase and regions of low salinity correspond to ice domains. We see that one single mode develops given by the wave number \( \kappa_c \). Similar to the one-dimensional case, we see the formation of one dominant wavelength also in two dimensions. This can be understood as the maximum of unstable wavelengths (11) which becomes

\[
\kappa_c = \psi_0 \frac{(D - 1)^2}{(D + 1)} \left[ (D - 1)(1 - 2\alpha_3 \psi_0) - 2\psi_0 + \frac{(D + 1)\psi_0^{1/2}\sqrt{(D - 1)(2\alpha_3 \psi_0 - 1) + \psi_0}}{\sqrt{D}} \right].
\]
The critical wave number sets the length scale on which phase separation occurs and is visible as the dominating coarse graining mode in fig. 3. The size of the solidification structures depends on the super-cooling relative to the freezing temperature $T_c$. The higher the super-cooling, the more rapidly water freezes and the smaller the structures become.

7 Comparison with experiments

Concerning the experiments, we suggest three types of comparisons: i) morphology, ii) percolation threshold and iii) structure size where our model describes realistic parameters. We will start with the morphology. For the web of brine channels one observes different textures, for instance granular ice, columnar-granular structures or plate ice. Figure 4(b) shows a measurement yielding granular texture [20] without prevalent orientation. In fig. 4(c) we have chosen the best fit of the former Turing model [26] to the structure size. If we compare with the simulation of our phase field model in fig. 4(d), the texture of the cast of brine channels seems to be better described by our present model than by the Turing model. Though the absolute size is not so much different, the three parameters of the Turing model had been adjusted to fit the structure as best as possible. Here, with the phase field model, we have chosen parameters according to the thermodynamic properties of water and have obtained the structure as a consequence of these parameters.

The structure of the brine pore space of single crystals [16] is shown in the iso-surface plots in fig. 4(a). The upper images clearly show near-parallel intra-crystalline brine layers. The view across the layers (bottom images) show brine layer textures much more complicated than suggested by the simple model of parallel ice lamellae and parallel brine sheets illustrated in fig. 4(b). Depending on the temperature, the images show a brine pore porosity from $p = 2.2\%$ to $8.8\%$. The connectivity increases with porosity as the pore space changes from isolated brine inclusions at $p = 2.2\%$ to extended, near-parallel layers at $p = 8.8\%$. The thermal evolution of the brine pore space with percolation theory was characterized in [32] where a connectivity threshold was found at a critical volume fraction $p_c = 4.6\%$. Below $p_c$ there are no percolating pathways spanning through the sample, i.e. the brine is trapped within the intra-crystalline brine layers.

Let us quantify this statement by a cluster size analysis of fig. 4(a) where the corresponding histograms are given in fig. 5. As one can find, the percolation transition is visible around 44% filling in the range of ($-8^\circ C$, $-4^\circ C$). Now we compare with our simulation varying the parameter $\alpha_3 = 0.9$ and $\alpha_3 = 1$. We see that the same per-
has a realistic description of seawater at 0.032 K supercooling and a lower limit of the supercooling region of fresh water at \(-18.78 \, ^\circ\text{C}\). For this growth condition we obtain \(2\pi/\kappa_c = 4975.25\) as dimensionless structure size and using eq. (16) the critical domain size is \(\lambda_c = 198 \, \mu\text{m}\) in agreement with the observed values. Brine inclusions [20] have scales in the range 3–1000 \(\mu\text{m}\), where the average dimension is typically 200 \(\mu\text{m}\).

8 Summary

To summarize, a model for the formation of salty water channels (brine entrapment) in sea ice has been developed which consists of two coupled order parameters, the tetrahedricity and the salinity preserving the mass conservation of salinity. The linear stability analysis provides a phase diagram in terms of two model parameters indicating the region where spatial structures can be formed due to the instability of the uniformly ordered phase. The region of instability is determined exclusively by the freezing parameter and the (specific heat) structure parameter and not by the diffusivity as it was the case in the reaction-diffusion Turing model [26]. This allows to link these model parameters to thermodynamical properties of water like super-heating, super-cooling, freezing temperature and specific heat simultaneously.

With the help of these model parameters, we solve the time-dependent coupled evolution equations and find a brine channel texture in agreement with the experimental values. That the physical justification of the parameters by other properties of water leads here to a better description of the brine channel texture, we attribute to the mass conservation invoked in the present model.

The presented model does not include yet the heat transfer. We have merely concentrated on structure formation at short time scales to consider the processes ab initio. Therefore the model should be extended to include the temperature field as a third order parameter. The inclusion of a velocity field is also necessary in order to describe real situations since convective motions certainly are expected to be present.

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Author contribution statement

The model set-up and linear stability analysis has been performed by all authors. The relation of model parameters to properties of water has been derived by S. Thoms and B. Kutschan. Picture analysis and histograms have been provided by K. Morawetz. Numerical codes were performed by B. Kutschan.
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