

Mathematics of Crystal Growth

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Abstract. For growing crystals, we introduce the effect function to describe the interaction among each other and we assume that a certain operator operating on the status function express the growth rate. On the basis of observation made by Nakaya *et al.*, we determine the class of the operator, who is necessary to give the dendritic growth of snow crystal. We conclude that, in order to have the dendritic growth in snow crystals, a nonlinear term is necessary beside the Laplacian on the status function, which may correspond to the diffusion efficiency.

1. Introduction

We propose a new mathematical method to read out physical structure from the information given through figures of phenomena.

As an application of the method, we investigate the dendritic growth of the snow crystal in the plane and determine the necessary type of non lineality of the differential equation describing the growth.

2. Preliminaries

We prepare several notations and develop the method on the plane Q .

A figure F in Q is a smooth domain in Q with a smooth tubular neighborhood. The boundary of a figure F is denoted by $\text{bd}(F)$ and the growth rate function w is a function defined on $\text{bd}(F)$ describing the growing speed of the figure in the normal direction $n(P)$:

$$w = dn/dt.$$

The characteristic function $ch(F)(P)$ of the figure F is a square integrable function given by:

$$ch(F)(P) = \begin{cases} 0, & \text{if } P \text{ is not in } F \\ 1, & \text{if } P \text{ is in } F \end{cases}$$

The following is our framework to develop mathematical theory of the figures of crystals.

We represent a crystal in Q by a pair (F, s) of a figure F in Q and a status function s on Q which is a square integrable function on the position P and the time t . The function $ch(F)(P)$ may be regarded as to represent the initial stage of the figure in the space of the square integrable functions.

We assume first that the status function s of a crystal F determines the growth rate w by a certain operator D such that

$$D(s) = w \text{ (approximately),}$$

the meaning of “approximately” will be clarified later.

We assume next that the status function is deformed by the interaction between the crystals F, F' , which is expressed by a function $E(F, F')$ of position P and the time t determined by $s(F)$ and $s(F')$ satisfying that:

$$ds/dt = E(F, F'), s(F)(P, 0) = ch(F)(P) \text{ (approximately),}$$

for the crystals F' other than F .

The meaning of the “approximate” equality is as follows:

For a neighborhood U of the origin in the real line R , the set of U -mollifiers is the set of the real non negative smooth functions $g(x)$ on R such that

- 1) the support is contained in U
- 2) invariant under the symmetry around the origin
- 3) the integration over the reals is the unity

$$\int f = 1$$

4) the k -th differential $g^{(k)}$ has k transversal intersections with the X -axis in U

The mollifier on R can be generalized naturally to that over Q by taking the distance between the point and the origin in Q in place of the variable x on R . For a symmetric neighborhood U in R of the origin, the disk of the radius U is also denoted by U .

For a square integrable function f on Q , the smoothing by a mollifier g is given by the convolution $g*f$

$$f*f(x) = \int g(y)f(x - y)dy$$

A property P for real square integrable functions is called mollifier invariant over U , if the smoothing $g*f$ satisfies P for any U -mollifier g and a property P is mollifier invariant if the property P is invariant over U , for any U in some neighborhood V .

For a neighborhood W of the identity in the space of diffeomorphisms of the plane with the infinitely differentiable topology, figures F, F' are called W -equivalent if there is a diffeomorphism h in W which sends F onto F' .

We can prove that Lemma. For a given neighborhood W of the identity in the space of diffeomorphisms of the plane, there exist a neighborhood U of the origin so that if a smooth function s yields a figure F from B by the equation

$$w = D(s)$$

then for any U mollifier g it holds that the figure F' generated from B by

$$w = D(g*s)$$

is W -equivalent to F .

Since a diffeomorphism near to the identity in the topology above does not change the “shape” in Q generically, we see that the small change in the choice of the mollifier is covered by the small diffeomorphism so that the shape is kept fixed generically, this may give a reasoning for the terminology “approximate”.

3. General Theory for Determination of Operators

For a constant c , the property that w or its differential is greater than or less than c is a mollifier invariant property, because a diffeomorphism near to the identity does not affect so much on w itself or its differential.

As is noticed by Barankin (1964), the actual observation may be done on this basis, that is, a constant growth is almost neglected in the observation of the growing shape and the main interest is that the growth is positive or negative with respect to the constant growth, which we may refer module constant reduction.

A mollifier invariant property is called universal when it is satisfied by all the translations of crystals, independent of its shape, of position.

Because theories in physics are established on the basis of smoothness of the phenomenon, we may assume that mathematical phenomenon can be physical law governing phenomenon has to be universal at any point, we may assume that the differential operator who describes the growth rate w of the crystal is universal.

In this framework, we investigate the growing process of a crystal which is described as follows:

First, a seed crystal of the shape $B = B(0)$ is given and is regarded as a mathematical crystal together with the characteristic function $ch(B(0))$ as the status function $s(0)$, then according to the growth rate function $w = D(s(0))$ it gives a new figure $B(1)$ after a small time interval ϵ . On the other hand, the effect function E acting on $s(0)$ gives the next status function $s(1)$ over $B(1)$, yielding the second crystal $(B(1), s(1))$, and so on.

Thus, the problem turns to read out the universal differential operators D and E from the information given through the growing figure of the crystal, especially those of the dendritic growth, on the basis of the reduction modulo constant.

For a technical reason, we assume that the differential operators D and E are of polynomial type, that is, we assume D and E are elements of the algebra V defined as follows: Denote by X, Y the differential operators

$$X(f) = df/dx, Y(f) = df/dy,$$

respectively, and denote by $*$ the multiplication given by the composition of the operators. The multiplication $*$ satisfies the distribution law with the conventional multiplication by the value. The polynomial algebra with respect to the conventional multiplication

$$V = V[X^p * Y^q], (p, q = 0, 1, 2, \dots)$$

over the monomials $X^p * Y^q$ ($p, q = 0, 1, 2, \dots$) of the operators X, Y turns out to be the algebra of differential operators, where X^p, Y^q stands for

$$X^p = X * X * \dots * X \text{ (p-times),}$$

$$Y^q = Y * Y * \dots * Y \text{ (q-times),}$$

respectively.

Now, we look for the operators D and E in the space V for the dendritic growth of the snow crystal growth.

We can show first that both of the operators should be a function of at most second order derivatives in x and y . In fact, suppose that there exists another crystal C in a neighborhood of B , then the effect of the crystal C on the growth of the crystal B is known to be always non positive modulo constant except when C is in a very thin layer around B independent of their shape as is noticed by Nakaya (1951). Therefore, $D(g)$ -c can change its sign only once around the edge $bd(B)$ of B for a sufficiently large class of the mollifiers g , yielding that the operator D is a function of at most second order derivatives.

We determine then a class of first and second operators in space V which

satisfies the independent condition of the universality.

Since the independence of the choice of the coordinate is equivalent to the invariance under the action of the orthogonal group, we can derive that the invariant differential operator in first and second order derivatives is a function $H(L, \det, \text{grad})$, of the Laplacian L and of differential operators \det and grad given by

$$\det(f) = (f_{xy})^2 - 4(f_{xx})(f_{yy}),$$

$$\text{grad}(f) = (f_x)^2 + (f_y)^2.$$

We see that the operators \det and grad change the sign more than once on the axis modulo constant for some mollifier, because they are of even degree in X and/or in Y , hence they can not be the variables in H . Thus, we may conclude that the growth rate operator D and the effect operator E and polynomial functions $H(L)$ and $E(L)$ of the Laplacian L , respectively, here the growth rate operator acts on the status function $s(B)$ to yield the growth rate $w(B)$ and the effect operator $E(L)$ acts on the status functions $s(B), s(C)$ to yield the effect function $E(B, C)$ of C on $\text{bd}(B)$.

For further determination of the effect operator, we assume again from experimental reason that it is additive, then we see that the possible type of operators is reduced to only a linear function K of the Laplacian L . Hence we have that

$$K(L) = aL + b, \text{ for constants } a, b$$

$$E(B, C)(P, t) = K(L)(s(B) + s(C))(P, t).$$

4. Determination of Operators for Dendritic Growth

In order to investigate the necessary types of the polynomial functions H and K for the dendritic growth, we fix notation for the growing process: Suppose we are given a seed crystal of shape B , then we take the pair $(B(0), s(0))$ of the shape $B(0) = B$ and the status function $s(0) = g^*ch(B)$ given by the (mollified) characteristic function as the mathematical translation of the seed crystal

$$\text{seed crystal} \rightarrow (B(0), s(0)), \text{ where } B(0) = B, s(0) = g^*ch(B),$$

The pair $(B(0), s(0))$ grows into next crystal $(B(1), s(1))$ after a short time unit e , where the shape $B(1)$ is determined by the end points of the normal vectors to $\text{bd}(B(0))$ of the length $eH(L)(s(0))$, which may be considered as a result of

attaching of $C(0)$ to $B(0)$,

$$B(1) = B(0) + C(0)$$

and the status function $s(1)$ is determined by

$$s(1)(P) = s(0)(P) + eK(L)(s(0) + g^*ch(C(0))).$$

This process is summarized in the following diagram

$$(B(0), s(0)) \rightarrow H(L) \rightarrow B(1)$$

$$B(1) \rightarrow K(L) \rightarrow s(1) \Rightarrow (B(1), s(1)),$$

thus, we have $(B(i), s(i))$ successively for $i = 1, 2, \dots$ by the following inductive relations:

$$B(i + 1) = \text{end points of normals to } bd(B(i)) \text{ of length } eH(L)(s(i))$$

$$B(i + 1) = B(i) + C(i)$$

$$s(i + 1)(P) = s(i)(P) + eK(L)(s(i) + g^*ch(C(i))).$$

With the above knowledge, we discuss the type of equations for the dendritic growth under several assumptions which may eventually be made clear by further experiments. Suppose we sume i is topologically equivalent to the length of the needle and the growth rate $H(L)s(i)$ is monotone in i on each side of the needle. This situation may be expressed as codimension zero catastrophe, thus we see that higher catastrophe is necessary for the dendritic growth.

On the other hand, the function $s(i)(P)$ increases first and after maximum decreases until the end of the needle as a quadratic polynomial in i generically, and its Laplacian behaves in another way, it increases after decrease, provided that the needle growth is not very quick. A rough reasoning is as follows. The equation for the status function is nothing but the heat equation in the limit, because $K(L)$ is equivalent to the Laplacian itself, therefore, we can use the fundamental solution $u(t, x)$ of the heat equation to investigate the process, because it is a successive attaching of (abstract) unit of crystals of a certain temperature, where unit of crystals means the growth aggregate of single crystals in a unit time. We see that $s(i)$ and the second derivative of $s(i)$ in P is a sum of the terms

$$2ve u(je, ve(j - x)),$$

and

$$-v^3 e^{2j-1} u(je, ve(j-x)) \text{ (approximately)}$$

for $j = 1$ to i , respectively, where v is the growth rate in needle direction, assumed to be constant and small. Thus, the first derivative is monotone and positive at one end and negative at the other end of the needle. By a similar argument for the second derivative, we have the assertion for the Laplacian.

Observation tells us that both needle and dendrite grows simultaneously, therefore, in order to have a dendritic growth, $H(L)s(i)$ has to increase first towards a maximum, decrease next towards a minimum and increase again till the end of the needle, so as to give a dendrite at the maximum point and suppressed growth at the minimum point. This situation may be realized generically by a third order polynomial in i , corresponding to the catastrophe of codimension one.

Hence, we see that the growth rate $H(L)s$ for dendritic growth is hardly realized by a linear function of the Laplacian $L(s)$, which is of second order in a generic sense and can be monotone or have only one minimum at most.

5. Conclusion and Remarks

Suppose that the generic argument is proved by the experiments, then we conclude that the growth rate operators consisting only of higher order term of the Laplacian $L(s)$ or of the status function s are not realistic for the dendritic growth, and that the possible operators must be only these equivalent to that containing the Laplacian $L(s)$ multiplied by a term containing s .

In fact, we see from above that a polynomial of order n of the status function or of the Laplacian is a polynomial of order $2n$ as a polynomial in i in a generic sense, doubling the number of the minima and the maxima, where dendrites grow. Thus, if we assume that the dendritic growth yields one dendrite at one time, the operators consisting only of the status function or of the Laplacian are impossible, because there results more than one dendrite at one time. Hence we see that the growth rate operator should necessarily contain both the Laplacian and the status function s . This is obvious except the case when it is expressed as a sum $P(L(s)) + Q(s)$ of polynomials P, Q in the Laplacian $L(s)$ and the status function s , respectively. If it is of the form $P(L(s)) + Q(s)$, we consider an operator $(1 + rP(L(s)))(1 + rQ(s))$ of multiplication form for a small r , which is equivalent to $P(L(s)) + Q(s)$ in the observation, that is, equivalent module constant and relatively small growth.

In connection with the terminology of mechanism, we may consider that the Laplacian corresponds to the diffusion process and the multiplication by the term of s to the diffusion efficiency in a generalized sense. In this setting, we can express the conclusion above as follows:

A diffusion process together with the diffusion efficiency plays the essential role to give the dendritic growth.

We admit the present note lacks precise experimental data which is now theoretically necessary. We hope to publish a completed theory in near future with further applications in a deduction of Shroedinger equation in quantum mechanics from the information on the shape, for example, using the parallelism in the effect function.

Acknowledgements

For the profound philosophy on mathematics and mechanism which motivated the present study, we owe the conversations with Professors E. Barankin and R. Thom. We are also grateful for the valuable discussions and for the suggestions during the preparation of this paper by Professors K. Aomoto, T. Hida, T. Kubota, and W. Shih from mathematical side and by Professors K. Higuchi, H. Mihashi, A. Ono, H. Tanaka, S. Fukui, S. Hayakawa, and R. Takagi from physical side, the constant encouragements by A. Ono that enabled this work.

REFERENCES

- Barankin, E. (1964), Probability and the east, *Ann. of the Inst. of Statistical Math.*, **16**, 185–230.
 Higuchi, K. (1961), On shape of ice crystals (in Japanese), *Kishoshushi*, **139**, 237–248.
 Komabayashi, M. (1972), Two dimensional computation of shape of an ice crystal growing in air, *J. Rech. Atoms.*, **16**, 307–328.
 Kuroda, T. (1982), Growth kinetics of ice single crystal from vapor phase and variation of its growth form, *J. Met. Soc. Jpn.*, **160**, 520–534.
 Nakaya, U. (1951), The formation of ice crystals, *Compendium of Met.*, 207–220.
 Shikata, A. and Shikata, Y., On a computer simulation of snow crystal growth (in prep.).
 Thom, R. (1972), *Stabilite structurelle et morphogenese*, Ben-jamin, Inc.