LOCAL TRANSFORMATIONS TO SIMULATE TWO DIMENSIONAL DENDRITIC CRYSTAL GROWTH. (*)

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Abstract.--Natural processes that are far from near-quilibrium and show forms of self- organization are extremely difficult to model with classical physical methods. Local elementary transformations in the sense of cellular-automata open an alternate view at the modelling of these processes.

INTRODUCTION

Many processes in nature occur in a thermodynamic state that is far from near-equilibrium. From a physical point of view, they are extremly difficult to model. Crystal growth of ice or the solidification of a molten metallic alloy are already too complex to be treated rigorously. In addition ice shows the full scale of growth forms from facet formation on crystallographic planes in depth hoar to the dendritic growth of crystals in supercooled water or in the free atmosphere. These examples - and of course any one from biology - are illustrations of highly non- equilibrium, self- organizing phenomena. According to Langer's (1980) excellent review on the classical state of pattern formation in crystal growth, to deal with self- organizing systems has become a fashionable occupation among physicists, chemists and mathematicians. The classical tools of mathematical physics, however, are rather inadequate to express the underlying processes, the related microscopic pattern and the resulting macroscopic shapes. To overcome this drawback, computer based methods have been used widely.

These actual trends may be characterized by the words of Maddox (1986): "Computer simulations of aggregation processes are fashionable because the problems are complicated. ... This fashion is a reaction to earlier disappointments, and particularly to the recognition that earlier macroscopic models of the growth of aggregates, which distinguished between the various positions on an extending surface only by their macroscopic properties and especially their curvature, have given only poor accounts of what really happens. ...". Physics seems to have degraded to fashion!

DENDRITIC CRYSTAL GROWTH, CLASSICAL APPROACH

In dendritic growth of ice crystals, the growth mechanisms are controlled by diffusion fields. The rate of growth is determined by the speed at which latent heat from the crystallization process is dissipated. In supercooled water, the diffusion of latent crystallization heat is the dominant part. In the free atmosphere the concentration of water vapor (molecules and droplets) and the concentration- and temperature gradients play an important role.

In order to have sidebranches forming on a growing dendrite, non- linear instabilities have to develop and have to be sustained by an appropriate mechanism. The marginal stability hypothesis by Langer (1980) includes surface tension and assumes that the speed at which the tip of a dendrite grows is proportional to the square of its curvature. In his paper's summary, his critical questions anticipate the findings of Honjo and Sawada (1985). With ammonium chloride growing in a quasi two dimensional set up, they have shown that this is not the case. How far then is the theory correct?

CRYSTAL GROWTH, ALTERNATE APPROACHES

Because of the impossiblity to rigorously formulate the growth processes, many authors have tried to simulate crystal growth with various numerical computing techniques (Kessler, 1984; Ben-Jacob et al.,1984). In diffusion limited aggregation (Sander, 1984) for instance, a particle diffuses from the outside to eventually hit a central seed and to stick there. The diffusion path is computed on a random walk basis in a square lattice. The growth is limited by the condition that the random walker has to hit a particle from a previous step.

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The cellular automata formalism, however, starts from the center towards the outside. A particle is incorporated in the frame if the conditions in the nearest neighborhood are favorable. This neighborhood is represented by a local, hexagonal coordinate frame. The growth algorithm can be combined with a Boolean function to switch between different growth regimes or with a random function to generate flaws and irregular shapes.

CELLULAR AUTOMATA FORMALISM

J. v. Neumann (1966) invented the cellular automaton and did the basic work in trying to simulate mechanisms of self reproducing systems in biology. Another biological analogon is the famous game of life, devised by J. H. Conway as a play on a checker board long before personal computers were available. Local selection rules in a Moore-coordinate frame (3*3) decide whether the central "particle" comes into existence, stays alive or is going to die (Hayes, 1984). The resulting patterns are very similar to actual cell cultures on a nutritive substrate. A systematic classification of one- dimensional cellular automata is to be found in the comprehensive review of Wolfram (1983).

Local transformations in a hexagonal coordinate frame

Under normal vapor pressure and temperature conditions, water crystallizes as ice I(h). Projection of oxygen molecules on a plane perpendicular to the crystallographic c- axis reveals a hexagonal structure of the ice skeleton. Non- equilibrium growth occurs preferentially in this plane. A hexagon is therefore a rough but reasonable approximation of an elementary structural element. It has to be mapped on a pseudo- hexagonal coordinate system, however, the computer arrays being inherently either linear or rectangular. For the central coordinate at the intersection of row i with column j, the "nearest neighbors" are located at i-1,j-1; i-1,j+1 for the previous row, at i,j-2; i,j+2 in the same row and for the row below at i+1,j-1; i+1,j+1. This coordinate frame has been used in this F77-program (appendix).

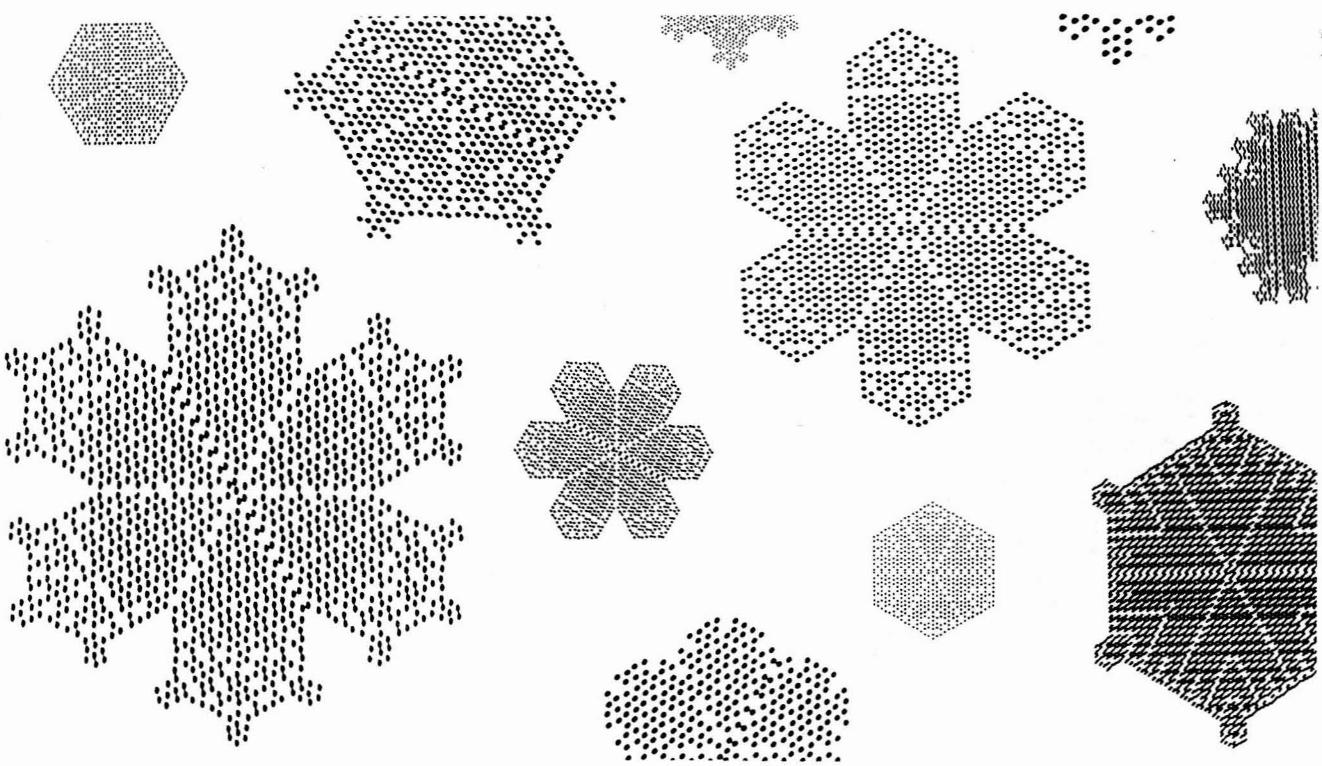

Figure 1. Simple XOR cellullar automaton resembling near- equilibrium hexagonal snow plates (from Good, 1985).

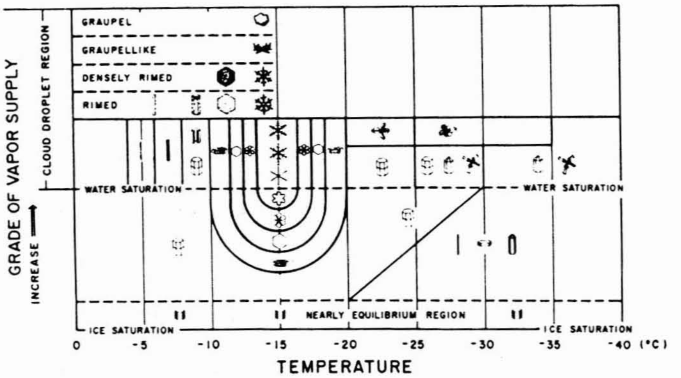

Figure 2. Magono and Lee (1966) diagram, vapor pressure over ice and water versus negative air temperature.

Two- dimensional, hexagonal XOR automaton

The algorithm used considers for each point within its six nearest neighbors, whether one and only one neighbor is "turned on". If this is true, the center of the coordinate frame switches from 0 to 1 (void -> ice). This corresponds to a logical exclusive OR (XOR) operation. After a number of steps, including the start of side branching, simple hexagons result. This is in fact the only way, "accretion" can initiate from a single point (nucleus) that is in its ON state. If the XOR algorithm is modified such that the transition 0 -> 1 can occur with one OR two neighbors turned on, no sidebranching is initiated and only pure hexagons result. In the XOR formalism, however, the growth of a new facet starts always from corner points. Figure 1 (Good, 1985) illustrates a selection of the resulting patterns. The computer algorithm performing this growth is to be found in the listing of the F77 program of the appendix.

Nature produces similar hexagons in the free atmosphere at -15 C and for a water vapor pressure between ice saturation and water saturation (Magono and Lee, 1966; Nakaya, 1954).

It is to be noted that in spite of the suggestion of the Magono- diagram, all shapes of figure 1 are equivalent having been generated by the same algorithm.

Dendritic growth from a hexagonal XOR automaton

In the free atmosphere, going to even stronger non-equilibrium conditions with faster growth rates (higher supersaturation), the facets cannot catch up with the dendrites that start from the six corners because the latent heat of crystallization is removed more easily from the tips of the dendrites.

The program simulates the (hexagonal) dendritic growth regime by ANDing the XOR- with the hexagonal direction formalism. A one-dimensional ray results. Changing to the undisturbed XOR algorithm from above to simulate the growth of near equilibrium forms, the needles will thicken and take on the shapes known from figure 1. By switching back and forth from hexagonal (XOR) growth to the dendritic (XOR AND hexagonal direction) growth, the dendrites extend in two dimensions.

Physically, the dendritic growth regime favors the outflow of the latent heat of crystallization by maximizing the active surface of the crystal. The rate of accretion may slow down by lowering the concentration of water molecules.

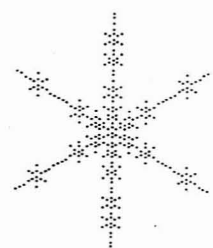

Figure 3. Hexagonal XOR dendrite. Long pure dendritic, short undisturbed growth periods.

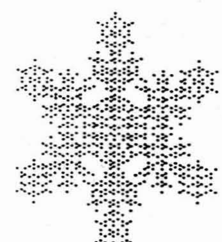

Figure 4. Hexagonal XOR dendrite with longer periods of undisturbed "near- equilibrium" growth than in figures 3.

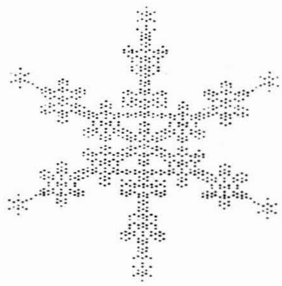

Figure 5. Hexagonal XOR dendrite. The vertical branches show restricted growth of the "undisturbed" hexagons (see figure 7)

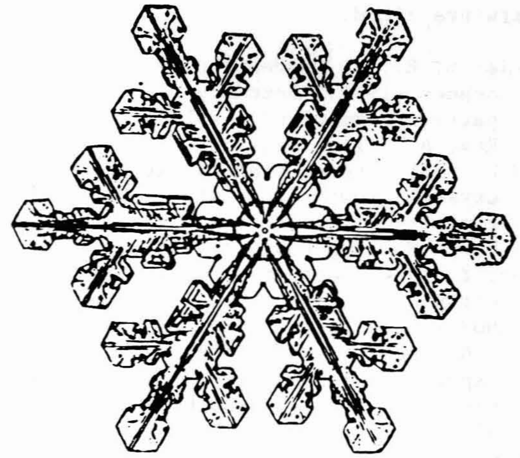

Figure 7. Natural dendritic snow crystal. Because of latent heat dissipation, only outward looking subbranches can grow.

In the diagram of Magono and Lee (1966) we would move downward, along a vertical line (T = - 15 C ; const) (figure 2) and the growth regime of near equilibrium shapes would result. Moving up and down in several cycles will produce a wide variety of dendritic shapes. Because the latent heat can best be dissipated at the outer boundary of the crystal, only the outward looking "subbranches" are stable in nature (figure 7).

The program does not account for this asymmetry, therefore symmetric two- dimensional shapes result on the needles. The simple figure 3 and the more complex one of figure 4 are illustrations of the algorithm. The truncation in the vertical branches of figure 5 is an artifact due to the limiting of the the vertical growth area by the parameter VB (Appendix).

CONCLUSION

The simple F77 algorithm in the appendix simulates only two distinct growth formalisms which are best illustrated by figures 1 and 6. Any gradual difference or asymmetry have yet to be introduced.

The aim of this paper, however, is not to present "physical" reality but to discuss an alternate approach to the still open problem of non- equilibrium crystal growth in the free atmosphere. The algorithm is strictly two- dimensional and does neither take into account concentration gradients of water vapor nor temperature gradients due to the dissipation of latent crystallization heat. The model assumes classical self- similarity where patterns from elementary processes are also to be found in the resulting macroscopic shapes. Complexity then would not arise in the elementary processes but in the extremely large number of steps between the submicroscopic- and the macroscopic world.

In addition, this sketch may help to enter the fascinating world of local transformations and cellular automata in an aesthetically rewarding field.

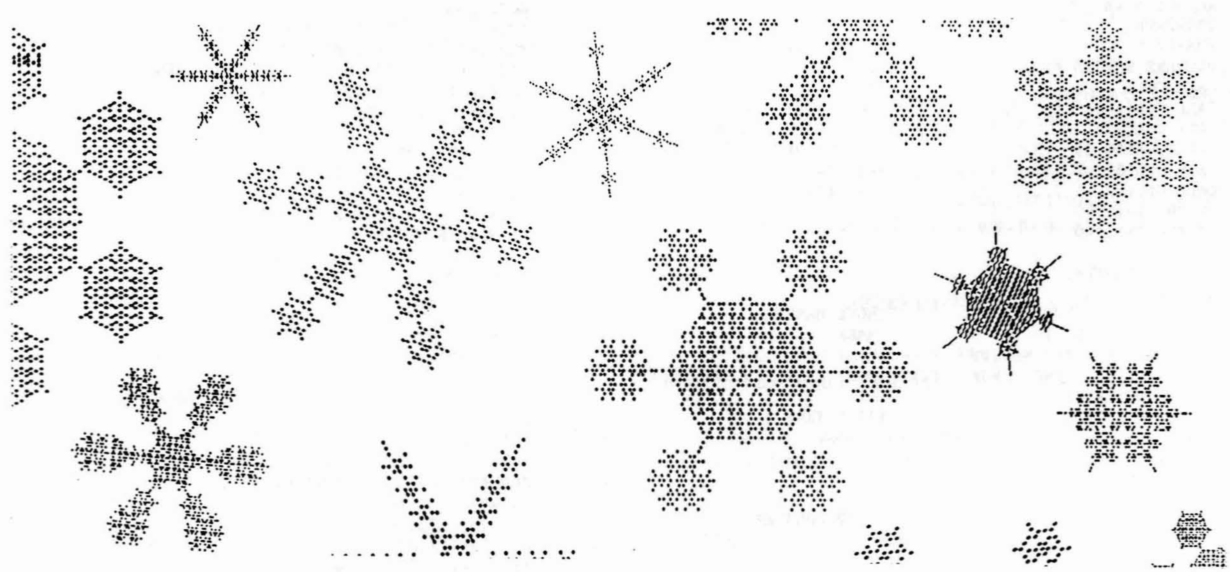

Figure 6. Collection of a few hexagonal XOR dendrites generated with the program listed in the appendix.

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APPENDIX

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C.......PROGRAM FLAKEDU.......FORTRAN-77........................
C.......GOOD...EISLF...WEISSFLUHJOCH...1985...1986..............
C.......SIMULATION OF HEXAGONAL GROWTH DIRECTIONS...............
        VIRTUAL B(140,140),A(140,140)   !2*19600 BYTES
        LOGICAL*1 A,B,Y,Z,ss
        INTEGER *2 C
        type *,' HEXAGONAL "FLAKES" WITH DENDRITIC REGIME'
        TYPE *,' GROWTH DIRECTIONS AND UNDISTURBED GROWTH'
        TYPE *,'  (SEE NAKAYA OR MAGONO AND LEE (1966))'
        TYPE *,'.................................'
        TYPE *,'         HEXAGONAL GROWTH UNTIL STEP'
        ACCEPT *,N1
        N1K-N1
        TYPE *,'         SHORT(1),MEDIUM(2), LONG(3) DENDRITES'
        ACCEPT *,11
        TYPE *,'         STEPS OF DENDRITIC GROWTH'
        ACCEPT *,IDG
        TYPE *,'         STEPS OF UNDISTURBED GROWTH'
        ACCEPT *,NSB
        ss-.true.
        Y-.true.
        DI-.5                  !DISCRIM. OR VIA ACCEPT
        SBR-0                  !OR VIA ACCEPT
        WD-0.0                 !OR VIA ACCEPT
C.......RANGE OF UNDISTURBED GROWTH.............................
        TYPE *,'         VERTICAL BANDWIDTH  (0,..DGR,UGR)'
        ACCEPT *,VB
        VBW-2*VB
        RVH--2                           !30,... DEGREES
C.......PREPARE SCREEN AND CGL..................................
        CALL CGL                         !INIT
        CALL CGL(103,'TI:',3)            !INIT VIEW SURFACE
        CALL CGL(92)                     !NEW FRAME (CLEAR)
        CALL CGL(86,1)                   !ORIGIN UPPER LEFT
        CALL CGL(80,.0,150.,.0,150.)     !WINDOW
        CALL CGL(82,.1,1.,.1,.9)         !VIEWPORT
        DO 200 I-1,140
        DO 200 J-1,140
200     A(I,J)-0
C.......SEED IN CENTER..........................................
        A(70,70)-1
        CALL CGL(1,70.,70.)              !MOVE MARKER
        CALL CGL(33,70.,70.)             !MARK
        TYPE 3,11,N1K,IDG,NSB,VB
3       FORMAT(6(/),' LONG   NGR   DGR   UGR   VBW'/I5,3I6,F6.1/)
C.......NUMBER OF LAYERS........................................
        DO 500 K-1,33                    !33 LAYERS
        IL-70-K
        IH-70+K
        JL-70-2*K
        JH-70+2*K
        II-0                             !STEP COUNTER
        DO 300 I-IL,IH
        II-II+1
        IJ-0                             !STEP COUNTER
        DO 300 J-JL,JH
        IJ-IJ+1
C.......AUTOMATON FOR HEXAGONAL SNOWFLAKE.........................
330     C-A(I-1,J-1)+A(I-1,J+1)+A(I,J-2)+A(I,J+2)+A(I+1,J-1)+A(I+1,J+1
350     B(I,J)-0
        IF(C.EQ.1)THEN                   !EXCLUSIVE OR
        IF(K.LE.N1K)GOTO 365             !SWITCH  0 ---> 1
C.......SELECTION OF GROWTH REGIMES...............................
        if(ss)THEN
        if(k.eq.(nl+idg))then
        nl-nl+nsb+idg
        ss-.FALSE.                       !switch to other regime
        endif
        ELSE
        if(k.eq.(nl+nsb))then
        nl-nl+idg+nsb
        ss-.TRUE.                        !switch again to other
        endif
        ENDIF
C.......DENDRITIC GROWTH..........................................
        ID-2*(K+1-II)
        JD-(2*K+1-IJ)
        RI-(2*K+1-IABS(ID))
        RJ-(2*K+1-IABS(JD))
        GOTO(351,352,353)LL
351     IF(.NOT.SS)GOTO 360
        GOTO 354
352     IF(K.LT.N1)GOTO 360
        GOTO 354
353     IF(K.LT.N1.AND..NOT.SS)GOTO 360
C.......SIMULATE GROWTH DIRECTIONS................................
354     IF(ID.EQ.0)GOTO 365              !VERTICAL
        AF-float(abs(JD))/float(abs(ID)) !HEXAGONAL
        IF((AF.LT.0.5).OR.(AF.GT.1.0))GOTO 300  !30,... (ASYM)
355     RR-1./RI
        IF(RJ.LT.RI)RR-1./RJ
        IF(RR.LT.DI)GOTO 300
        GOTO 365
360     IF(ABS(ID)-K.LT.SBR.AND.RJ.GT.VBW)GOTO 300
        RL-SQRT(RJ*RJ+RI*RI)
        IF((RL-K).GT.RVH)GOTO 365        !NON VERT.
        GOTO 300                         !SKIP
365     B(I,J)-1                         !SWITCH ON
C.......DISPLAY...................................................
        FI-FLOAT(I)
        FJ-FLOAT(J)
        CALL CGL(1,FI,FJ)                !MOVE POINTER
        CALL CGL(33,FI,FJ)               !DRAW POINT
        ENDIF
300     CONTINUE
        TYPE 1,K
1       FORMAT(21H+       STEPCOUNTER: ,I2)
        DO 400 I-IL,IH
        DO 400 J-JL,JH
400     A(I,J)-B(I,J)                    !COPY WORKPAGE
        IF (K.GT.N1K)VBW-VBW+1           !EXTEND VERTICAL RANGE
500     CONTINUE
        STOP 'FLAKE'
        END
```