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ERGODIC PROPERTIES OF CRYSTALLIZATION PROCESSES

Abstract. We consider a birth and growth process with germs being born according to a Poisson point process whose intensity measure is invariant under translations in space. The germs can be born in unoccupied space and then start growing until they occupy the available space. In this general framework, the crystallization process can be characterized by a random field which, for any point in the state space, assigns the first time at which this point is reached by a crystal. Under general conditions on the growth speed and geometrical shape of free crystals, we prove that the random field is mixing in the sense of ergodic theory. This result is illustrated by applications to the problem of parameter estimation.

1. Introduction

We consider the crystallization process which deals with points, called germs, $g = (x_g, t_g)$ in the space $\mathbb{R}^d \times \mathbb{R}^+$, where t_g denotes random time and x_g random location. The germ birth process $\mathcal{N}$ is a Poisson point process on $\mathbb{R}^d \times \mathbb{R}^+$ with intensity measure Λ. Once germs or crystallization centers are born, crystals grow if their location is not yet occupied by another crystal. When two crystals meet, the growth stops at the meeting point.

To describe crystal expansion in unoccupied space, for a germ $g = (x_g, t_g)$ and a point x in $\mathbb{R}^d$, let $A_g(x)$ be the time when the point x is reached by the crystal born in the location x_g at the time t_g. The crystallization process is then characterized by the random field (r.f.) ξ, which, for any location x in $\mathbb{R}^d$, assigns its crystallization time

$$\xi(x) = \inf_{g \in \mathcal{N}} A_g(x).$$

Consequently, at time t, a free crystal is the set $C_g(t) = \{x \mid A_g(x) \le t\}$.

The above model was introduced by Kolmogorov [1] and, independently, by Johnson and Mehl [6]. It has been intensively studied by many authors, including Møller [2, 3], Micheletti & Capasso [7], who represent main approaches. In these publications one can also find exhaustive lists of references. A very large part of these investigations deals with geometrical structures of mosaics after all the germs have been grown. In contrast, our main attention in the current work is on ergodic properties of the crystallization process, thus providing a base for efficient estimation of model parameters and subsequent analysis of limit theorems such as asymptotical normality.

The rest of the paper is organized as follows. Under general assumptions, we state in Sec. 2 that the r.f. ξ is mixing in the sense of ergodic theory. The proof is reported to Sec. 4. In Sec. 3, we give two examples of application of Theorem 1 to the problem of parameter estimation.

2. Assumptions on the birth and growth process and mixing

Germs are born according to a Poisson point process $\mathcal{N}$ on $E = \mathbb{R}^d \times \mathbb{R}^+$. That is, germs are random points $g = (x_g, t_g)$ in E, where x_g is the location in the growth space $\mathbb{R}^d$ and t_g is the birth time on the time axis $\mathbb{R}^+$. We suppose that the intensity measure of $\mathcal{N}$ has the expression

$$\Lambda = \lambda^d \times m,$$

where λ^d is the Lebesgue measure on $\mathbb{R}^d$ and m is a measure on $\mathbb{R}^+$ such that $m([0,a]) < \infty$ for all $a > 0$. The cases to be considered below (cf. [2]) are those with a discrete measure m and with a density measure $m(dt) = \alpha t^{\beta-1}\lambda(dt)$, where α, $\beta > 0$ are parameters. Since the Lebesgue measure is invariant under translations on $\mathbb{R}^d$, we have that $\mathcal{N}$ is space homogeneous.

For time t, we consider the so called *causal cone* $K_t = \{g \in E \mid A_g(0) \leq t\}$, which consists of all possible germs that can reach the origin before t. The measure $\Lambda(K_t)$ of the causal cone K_t is denoted by $F(t)$. These set and function play important roles in the sequel.

We assume that, for any germ $g = (x_g, t_g)$, the associated free crystal at time $t \geq t_g$ is equal to $C_g(t) = x_g \bigoplus [V(t) - V(t_g)]K$, where K is a convex compact set such that $0 \in K^\circ$ with $\bigoplus$ denoting the Minkowski sum, and $V(t)$ is an absolutely continuous function of t whose value is the distance achieved with positive speed $v(t)$. Finally, let M be a constant such that $v \leq M$, and let D_K be the diameter of the smallest ball centered at zero and containing K. Note that when $K = B(0,1)$ and $v = M$, then we have the well-known model which corresponds to the linear expansion in all directions at a constant speed.

We next consider the mixing of the r.f. ξ. To start with, we assume without loss of generality that ξ is a canonical r.f. on $(\Omega, \mathcal{F}, \mathbb{P})$. Namely, we suppose that $\Omega = \mathbb{R}^T$ with $T = \mathbb{R}^d$, $\mathcal{F}$ is the σ-field generated by the cylinders, and $\mathbb{P}$ is the distribution of ξ so that for all $\omega \in \Omega$, $\xi(x,\omega) = \omega(x)$. Since the Lebesque measure on $\mathbb{R}^d$ is invariant under translations, the r.f. ξ is homogeneous, that is, $\mathbb{P}$ is invariant under the translations $S_h(\omega)(x) = \omega(x+h)$ for all h in $\mathbb{R}^d$. We say that the canonical r.f. is mixing if, for all A and $B \in \mathcal{F}$,

$$\mathbb{P}\{A \cap S_h^{-1}(B)\} \underset{|h|\to\infty}{\longrightarrow} \mathbb{P}\{A\}\mathbb{P}\{B\}. \tag{1}$$

Note that every mixing r.f. in the above sense is ergodic. We have the following theorem.

Theorem 1. *For $d \geq 1$, the r.f. $\xi = (\xi(x))_{x\in\mathbb{R}^d}$ is mixing.*

The proof of the theorem is contained in Sec. 4 while the next section is devoted to the applications of Theorem 1 to the problem of parameter estimation.

3. Parameters estimation of the intensity measure

We consider two cases:

A) The measure m which is a component of intensity measure Λ is absolutely continuous and $m(dt) = \alpha\, t^{\beta-1} dt$ with α, $\beta > 0$,

B) The measure m is discrete: $m = \sum_{i=1}^n p_i\, \delta_{a_i}$ with $\sum_{i=1}^n p_i = 1$, for all $i = 1, \dots, n$, $p_i > 0$ and $0 < a_1 < a_2 \cdots < a_n$.

Moreover, we suppose, keeping notations of the previous section, that the crystal's shapes are defined by the compact $K = B(0,1)$ and that the growth speed is constant $v = 1$.

First of all we remark that the marginal distribution function

$$\mathcal{F}(t) = \mathbb{P}\{\xi(0) \leq t\}, \quad t \in \mathbb{R}^+,$$

can be expressed as follows

$$\begin{aligned} \mathcal{F}(t) &= \mathbb{P}\{\mathcal{N} \cap K_t \neq \emptyset\} \\ &= 1 - \mathbb{P}\{\mathcal{N} \cap K_t = \emptyset\} \\ &= 1 - \exp(-\Lambda(K_t)), \end{aligned}$$

where $K_t = \{g \in E \mid A_g(0) \leq t\}$ is a causal cone.

Hence

$$\Lambda(K_t) = -\log(1 - \mathcal{F}(t)). \tag{2}$$

On the other side Theorem 1 shows that the function

$$\widehat{\mathcal{F}}_T(t) = \frac{1}{T^d} \int\limits_{[0,T]^d} 1_{[0,t]}(\xi(x))\lambda^d(dx)$$

is a consistent estimator for $\mathcal{F}$:

$$\widehat{\mathcal{F}}_T(t) \xrightarrow{p.s.} \mathbb{E}\left(1_{[0,t]}(\xi(0))\right) = \mathcal{F}(t). \tag{3}$$

Now, using (2) and (3), we will easily construct consistent estimators for parameters α, β in case **A)** as well as for p_i in case **B)**.

3.1. Absolutely continuous case

If $m(dt) = \alpha\, t^{\beta-1}dt$, we have for all $t \in \mathbb{R}^+$

$$\begin{aligned} \Lambda(K_t) &= \int\limits_0^t \lambda^d(B(0,t-s))\alpha\, s^{\beta-1}ds \\ &= c_d \int\limits_0^t (t-s)^d \alpha\, s^{\beta-1}ds \\ &= c_d\, \alpha\, t^{d+\beta}\, l_d(\beta), \end{aligned} \tag{4}$$

where

$$c_d = \lambda^d(B(0,1))$$

and

$$l_d(\beta) = \frac{d!}{\beta\,(\beta+1)\ldots(\beta+d)}.$$

From (2) and (4), we deduce for all $t \in \mathbb{R}^+$ that

$$-\log(1-\mathcal{F}(t)) = c_d\, \alpha\, t^{d+\beta}\, l_d(\beta). \tag{5}$$

Taking in (5) $t = t_1$ and $t = t_2$, we obtain the following system:

$$\begin{cases} \dfrac{\log\left(\frac{\log(1-\mathcal{F}(t_1))}{\log(1-\mathcal{F}(t_2))}\right)}{\log t_1 - \log t_2} - d = \beta \\ -\log(1-\mathcal{F}(t_1)) = c_d\, \alpha\, l_d(\beta)\, t_1^{d+\beta} \end{cases} \tag{6}$$

From (3), we get immediately

Proposition 1. *The following statistics are strongly consistent estimators for parameters α and β:*

$$\widehat{\beta}_T := \frac{\log\left(\frac{\log(1-\widehat{\mathcal{F}}_T(t_1))}{\log(1-\widehat{\mathcal{F}}_T(t_2))}\right)}{\log t_1 - \log t_2} - d \xrightarrow{p.s.} \beta$$

$$\widehat{\alpha}_T := \frac{-\log(1-\widehat{\mathcal{F}}_T(t_1))}{c_d\, l_d(\widehat{\beta}_T) t_1^{d+\widehat{\beta}_T}} \xrightarrow{p.s.} \alpha.$$

Naturally, for particular values of t_1, t_2 the formulas for $\widehat{\beta}_T$, $\widehat{\alpha}_T$ could be simplified. For example, if $t_1 = e$, $t_2 = 1$, then

$$\widehat{\beta}_T = \log\left(\frac{\log(1-\widehat{\mathcal{F}}_T(e))}{\log(1-\widehat{\mathcal{F}}_T(1))}\right) - d, \quad \widehat{\alpha}_T = \frac{-\log(1-\widehat{\mathcal{F}}_T(e))}{c_d\, l_d(\widehat{\beta}_T)}.$$

3.2. Case of the discrete measure

We suppose now that m is the discrete measure $\sum_{i=1}^n p_i\, \delta_{a_i}$ with $\sum_{i=1}^n p_i = 1$, $p_i > 0$ and $0 < a_1 < a_2 \cdots < a_n$. Moreover, as the crystals can be born only at the moment $a_i, i = 1, \ldots, n$, the estimation of these moments is not a difficult task, and we can assume that $a_i, i = 1 \ldots n$, are known.

For all $t \in \mathbb{R}^+$,

$$\Lambda(K_t) = c_d \sum_{i=1}^n p_i\, (t-a_i)^d 1_{\{a_i \le t\}}. \tag{7}$$

Thus, if we consider Eq. (7) for $t = a_i$ with $i = 2, \ldots, n$ and $t = a_1 + a_n$, we obtain the following system:

$$\begin{cases} -\log(1-\mathcal{F}(a_2)) &= c_d\left(p_1\,(a_2-a_1)^d\right) \\ -\log(1-\mathcal{F}(a_3)) &= c_d\left(p_1\,(a_3-a_1)^d + p_2\,(a_3-a_2)^d\right) \\ & \vdots \\ -\log(1-\mathcal{F}(a_n)) &= c_d \sum\limits_{i=1}^{n-1} p_i\,(a_n-a_i)^d \\ -\log(1-\mathcal{F}(a_1+a_n)) &= c_d\left(p_1 a_n^d + \sum\limits_{i=2}^{n-1} p_i\,(a_1+a_n-a_i)^d + p_n a_1^d\right). \end{cases}$$

Again from (3), we derive the following consistent estimators $\widehat{p}_{i,T}$ of p_i for $i = 1, \ldots, n$.

Proposition 2. *The following statistics are strongly consistent estimators for parameters p_i:*

$$\begin{aligned}
\widehat{p}_{1,T} &:= \tfrac{1}{(a_2-a_1)^d}\left(\tfrac{-\log(1-\widehat{\mathcal{F}}_T(a_2))}{c_d}\right),\\
\widehat{p}_{2,T} &:= \tfrac{1}{(a_3-a_2)^d}\left(\tfrac{-\log(1-\widehat{\mathcal{F}}_T(a_3))}{c_d} - \widehat{p}_{1,T}(a_3-a_1)^d\right),\\
&\vdots\\
\widehat{p}_{n-1,T} &:= \tfrac{1}{(a_n-a_{n-1})^d}\left(\tfrac{-\log(1-\widehat{\mathcal{F}}_T(a_n))}{c_d} - \sum_{i=1}^{n-2}\widehat{p}_{i,T}(a_n-a_i)^d\right),\\
\widehat{p}_{n,T} &:= \tfrac{1}{a_1^d}\left(\tfrac{-\log(1-\mathcal{F}(a_1+a_n))}{c_d} - \widehat{p}_{1,T}\,a_n^d - \sum_{i=2}^{n-1}\widehat{p}_{i,T}\,(a_1+a_n-a_i)^d\right).
\end{aligned}$$

4. Proof of Theorem 1

To prove that a random field is mixing, it is sufficient to verify condition (1) for cylinders and establish the following condition

$$\begin{aligned}
&\forall x_1,\dots,x_k,\quad \forall y_1,\dots,y_m,\quad \forall E_1\in\mathcal{B}^k,\quad \forall E_2\in\mathcal{B}^m\\
&\mathbb{P}\{(\xi(x_1),\dots,\xi(x_k))\in E_1,\ (\xi(y_1+h),\dots,\xi(y_m+h))\in E_2\}\\
&\underset{|h|\to\infty}{\longrightarrow}\ \mathbb{P}\{(\xi(x_1),\dots,\xi(x_k))\in E_1\}\mathbb{P}\{(\xi(y_1),\dots,\xi(y_m))\in E_2\}.
\end{aligned}\tag{8}$$

We need three auxiliary lemma.

Lemma 1. *If A_1, A_2, B_1, and B_2 are four events, then*

(i) $|\mathbb{P}(A_1)-\mathbb{P}(A_2)|\le\mathbb{P}(A_1\triangle A_2)$,

(ii) $|\mathbb{P}(A_1\cap B_1)-\mathbb{P}(A_2\cap B_2)|\le\mathbb{P}(A_1\triangle A_2)+\mathbb{P}(B_1\triangle B_2)$,

where for two events A and B, $A\triangle B=(A\cap B^c)\cup(A^c\cap B)$.

Proof. These facts are well known. □

Now for all $h\in\mathbb{R}^d$ and $r\ge 0$, we define new random fields to approximate $\xi(x)$ and its translations $\xi(x+h)$:

$$\xi_r^h(x)=\inf_{\substack{g\in\mathcal{N}\\ |x_g-h|\le r}} A_g(x).$$

Lemma 2. *Let* $H(R) = \sup_{|x|=R} A_{(0,R)}(x)$. *Under our assumptions we have for all* $h \in \mathbb{R}^d$

$$\mathbb{P}\left\{\xi(x+h) = \xi^h_{(M'+1)H(R)}(x),\ |x| \leq R\right\} \geq 1 - e^{-\Lambda(K_{0,R})},$$

where $M' = MD_K$ *and* $K_{0,R}$ *is the causal cone defined as follows*

$$K_{(0,R)} = \{g \in E,\ A_g(0) \leq R\}.$$

Proof. As $\mathcal{N}$ is space homogeneous,

$$\begin{aligned}&\mathbb{P}\left\{\xi(x+h) = \xi^h_{(M'+1)H(R)}(x),\ |x| \leq R\right\}\\ =&\mathbb{P}\left\{\xi(x) = \xi^0_{(M'+1)H(R)}(x),\ |x| \leq R\right\}\end{aligned}$$

and it is then sufficient to demonstrate Lemma 2 for $h = 0$.

It is not difficult to see that

$$\{\xi(0) \leq R\} \subset \left\{\sup_{|x|\leq R} \xi(x) \leq H(R)\right\}.$$

Now, let us prove that

$$\left\{\sup_{|x|\leq R} \xi(x) \leq H(R)\right\} \subset \{\xi(x) = \xi^0_{R+M'H(R)}(x), \quad |x| \leq R\}. \tag{9}$$

Assumptions on the growth of crystals imply that for all germ g,

$$A_g(x) \geq t_g + \frac{|x - x_g|}{M'} \quad \forall x \in \mathbb{R}^d.$$

In particular, for germs g such that $|x_g| > R + M'H(R)$, we deduce that

$$A_g(x) > H(R) \quad \forall x \in \mathbb{R}^d, |x| \leq R.$$

Hence, for all x such that $|x| \leq R$,

$$\inf_{\substack{g\in\mathcal{N}\\ |x_g|>R+M'H(R)}} A_g(x) > H(R) \geq \xi(x),$$

and (9) follows.

On the other hand, for $0 \leq r_1 \leq r_2$ and $\forall x \in \mathbb{R}^d$

$$\xi(x) \leq \xi^0_{r_2}(x) \leq \xi^0_{r_1}(x).$$

As $R \leq H(R)$, we deduce

$$\{\xi(x) = \xi_{R+M'H(R)}(x),\ |x| \leq R\} \subset \{\xi(x) = \xi_{(M'+1)H(R)}(x),\ |x| \leq R\}.$$

Finally, we get

$$\mathbb{P}\left\{\xi(x) = \xi_{(M'+1)H(R)}(x),\ |x| \leq R\right\} \geq \mathbb{P}\{\xi(0) \leq R\}$$

and

$$\mathbb{P}\{\xi(0) \leq R\} = \mathbb{P}\{\mathcal{N} \cap K_{0,R} \neq \emptyset\}.$$

But,

$$\mathbb{P}\{\mathcal{N} \cap K_{0,R} \neq \emptyset\} = 1 - e^{-\Lambda(K_{0,R})}.$$

□

Lemma 3. *Under our assumptions*

$$\Lambda(K_{0,R}) \underset{R\to\infty}{\longrightarrow} \infty.$$

Proof. The assumptions on the growth of crystals imply that for all germ $g \in E$, there exists $R > 0$ such that $g \in K_{(0,R)}$ or equivalently such that 0 belongs to the crystal $C_g(R)$. But,

$$\bigcup_{R\geq 0} K_{(0,R)} = E$$

and since $\Lambda(E) = +\infty$, the result follows. □

We come back to the proof of Theorem 1.

Proof. For $(x_1, \ldots, x_k)$ in E^k, $(y_1, \ldots, y_m)$ in E^m, $E_1 \in \mathcal{B}^k$ and $E_2 \in \mathcal{B}^m$, we define the sets:

$$\begin{aligned} A &= \{(\xi(x_1), \ldots, \xi(x_k)) \in E_1\}, \\ B &= \{(\xi(y_1), \ldots, \xi(y_m)) \in E_2\}, \\ B_h &= \{(\xi(y_1 + h), \ldots, \xi(y_m + h)) \in E_2\}. \end{aligned}$$

Let $\epsilon > 0$ and $r = \max\{|x_i|,\ i = 1 \dots k;\ |y_j|,\ j = 1 \dots m\}$. By Lemmas 2 and 3, we can find $R > r$ such that

$$\mathbb{P}\left\{\xi(x) = \xi^0_{(M'+1)H(R)}(x),\ |x| \leq R\right\} \geq 1 - \epsilon.$$

Let us now take $h \in \mathbb{R}^d$ such that $|h| > 2R_1$, where $R_1 = (M'+1)H(R)$, and introduce the sets:

$$\begin{aligned} \widetilde{A} &= \{(\xi^0_{R_1}(x_1), \dots, \xi^0_{R_1}(x_k)) \in E_1\}, \\ \widetilde{B} &= \{(\xi^0_{R_1}(y_1), \dots, \xi^0_{R_1}(y_m)) \in E_2\}, \\ \widetilde{B}_h &= \{(\xi^h_{R_1}(y_1), \dots, \xi^h_{R_1}(y_m)) \in E_2\}. \end{aligned}$$

By Lemma 1 (ii), we have

$$|\mathbb{P}(A \cap B_h) - \mathbb{P}(\widetilde{A} \cap \widetilde{B}_h)| \leq \mathbb{P}(A \triangle \widetilde{A}) + \mathbb{P}(B_h \triangle \widetilde{B}_h).$$

Let $D = \{\xi(x) = \xi^0_{R_1}(x),\ |x| \leq R\}$. Then, by Lemma 2,

$$\begin{aligned} \mathbb{P}(A \triangle \widetilde{A}) &= \mathbb{P}((A \triangle \widetilde{A}) \cap D) + \mathbb{P}((A \triangle \widetilde{A}) \cap D^c) \\ &= \mathbb{P}((A \triangle \widetilde{A}) \cap D^c) \\ &\leq \mathbb{P}(D^c) \\ &\leq \epsilon. \end{aligned}$$

Replacing the set D by $D_h = \{\xi(x+h) = \xi^h_{R_1}(x),\ |x| \leq R\}$, we obtain by the same arguments that

$$\mathbb{P}(B_h \triangle \widetilde{B}_h) \leq \epsilon.$$

These two inequalities imply that

$$|\mathbb{P}(A \cap B_h) - \mathbb{P}(\widetilde{A} \cap \widetilde{B}_h)| \leq 2\epsilon. \tag{10}$$

On the other hand, the events $\widetilde{A}$ and $\widetilde{B}_h$ are independent because $|h| > 2R_1$. Thus,

$$\mathbb{P}(\widetilde{A} \cap \widetilde{B}_h) = \mathbb{P}(\widetilde{A})\mathbb{P}(\widetilde{B}_h)$$

and by space homogeneity of $\mathcal{N}$, $\mathbb{P}(\widetilde{B}_h) = \mathbb{P}(\widetilde{B})$, so that

$$\mathbb{P}(\widetilde{A} \cap \widetilde{B}_h) = \mathbb{P}(\widetilde{A})\mathbb{P}(\widetilde{B}). \tag{11}$$

Moreover, by Lemma 1 (i),

$$|\mathbb{P}(A)\mathbb{P}(B) - \mathbb{P}(\widetilde{A})\mathbb{P}(\widetilde{B})| \le |\mathbb{P}(A) - \mathbb{P}(\widetilde{A})| + |\mathbb{P}(B) - \mathbb{P}(\widetilde{B})|$$
$$\le \mathbb{P}(A \triangle \widetilde{A}) + \mathbb{P}(B \triangle \widetilde{B}),$$

and Lemma 2 implies that

$$\mathbb{P}(A \triangle \widetilde{A}) + \mathbb{P}(B_h \triangle \widetilde{B}_h) \le 2\epsilon. \tag{12}$$

Inequalities (10), (11), and (12) imply that for all $h \in \mathbb{R}^d$ such that $|h| > 2R_1$,

$$|\mathbb{P}(A \cap B_h) - \mathbb{P}(A)\mathbb{P}(B)| \le 4\epsilon$$

and the Theorem 1 then proved. □

5. Concluding remarks

• The method used in the proof of Theorem 1 can be applied (after some modifications) to estimate the rate of decreasing of the absolute regularity coefficient (for details see [5]). These results provide a solid base for establishing of asymptotical normality of estimators of model parameters.

• In Sec. 3, one example of application of Theorem 1 to the problem of consistent estimation was given. There are many others, we can mention here the mean value of crystals born in unit volume or, more generally, another functional of additive character. We are preparing a more detailed publication in this subject.

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