Crystallization processes : ergodic properties and statistical inference

Joint work with Youri Davydov

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1. Crystallization model
   - Description
   - Assumptions

2. Ergodic properties
   - Ergodicity
   - β-mixing coefficients

3. Parameters estimation
   - Absolutely continuous case
   - Case of a discrete measure

- Germs: $g = (x_g, t_g) \in \mathbb{R}^d \times \mathbb{R}^+$
  - $x_g \in \mathbb{R}^d$ crystallization center location in the growth space
  - $t_g \in \mathbb{R}^+$ crystallisation center birth time
- Birth process: Poisson point process $\mathcal{N}$ on $\mathbb{R}^d \times \mathbb{R}^+$ with intensity measure:

$$\Lambda(dx \times dt) = \lambda^d(dx) \times m(dt)$$

  - λ^d Lebesque measure on $\mathbb{R}^d$
  - m locally finite measure on $\mathbb{R}^+$
- Crystals growth: Θ_t = Portion of $\mathbb{R}^d$crystallized at time t
  - If $x_g \in \Theta_{t_g}$: no crystal starts growing at x_g
  - If $x_g \notin \Theta_{t_g}$: instantaneous growth of a crystal at x_g (shape/speed to be defined)
  - Growth stops at the meeting points

Model intoduced by Kolmogorov (1937) and Johnson & Mehl (1939)

Dimension 2

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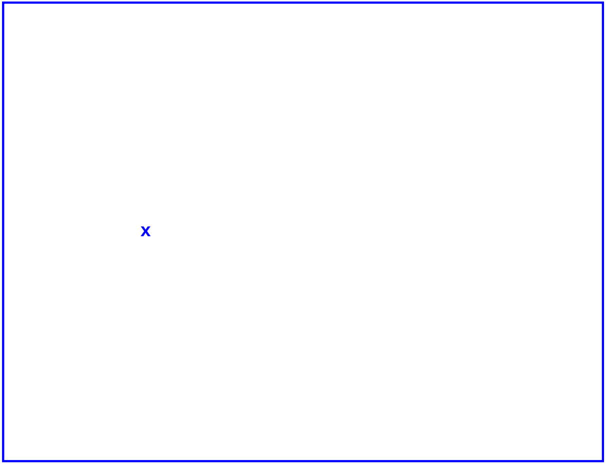

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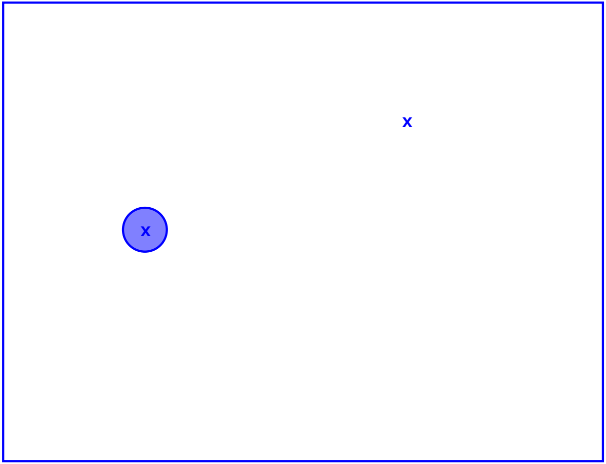

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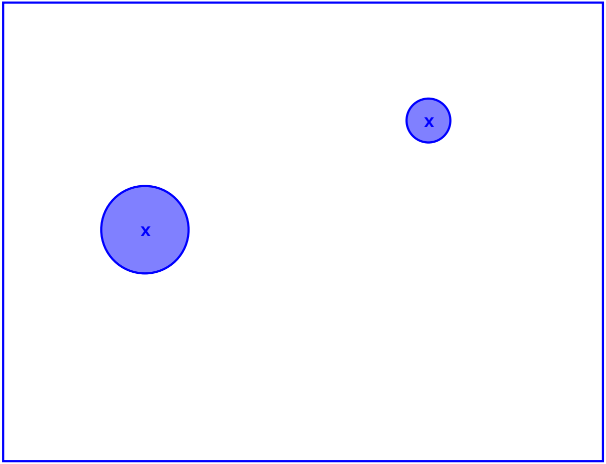

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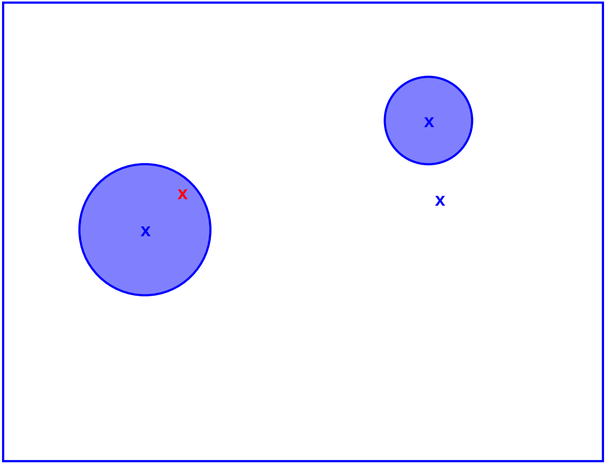

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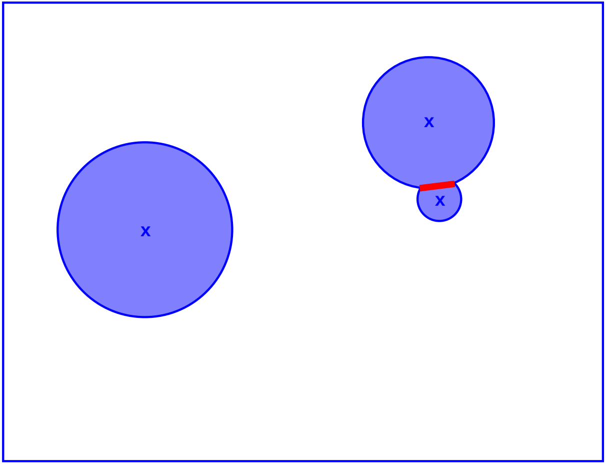

- Germination process:

$$\Theta_t = \text{Portion of } \mathbb{R}^d \text{crystallized at time } t$$

The set $\mathcal{N}_c$ of germs g_c giving birth to a crystal is a point process with intensity measure:

$$(1 - \mathbb{1}_{\Theta_{t-}})\Lambda(dx \times dt)$$

Capasso & Micheletti (1995,97...) approach

- Møller (1992,95...) approach:
  1. Assume, first, that all germs give birth to a crystal: the germination process is the Poisson point process denoted by $\mathcal{N}$ with intensity measure:

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Free crystal

A free crystal is a crystal which grows freely and originates from a germ born in a location not yet occupied by other crystals at the time of its birth ($x_g \notin \Theta_{t_g}$)

For all germ $g \in \mathbb{R}^d \times \mathbb{R}^+$,

- for all $x \in \mathbb{R}^d$, $A_g(x)$ is the *crystallization time* of x by the crystal associated to the germ g and assumed to be free
- for all $t \in \mathbb{R}^+$, $C_g(t) = \{x \in \mathbb{R}^d \mid A_g(x) \leq t\}$ is the *free crystal* associated to the germ g

Crystallization random field

For all $x \in \mathbb{R}^d$,

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

is the crystallization time of the location x. The crystallization process is then caracterized by the random field $(\xi(x))_{x \in \mathbb{R}^d}$

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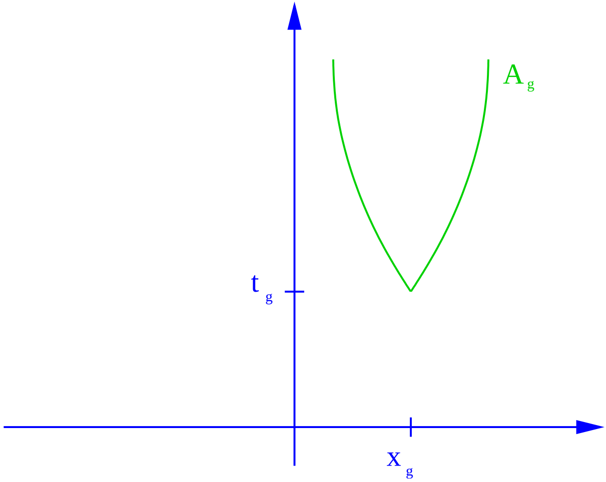

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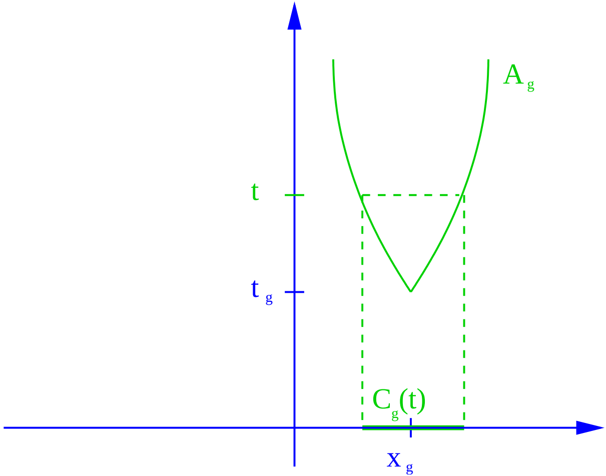

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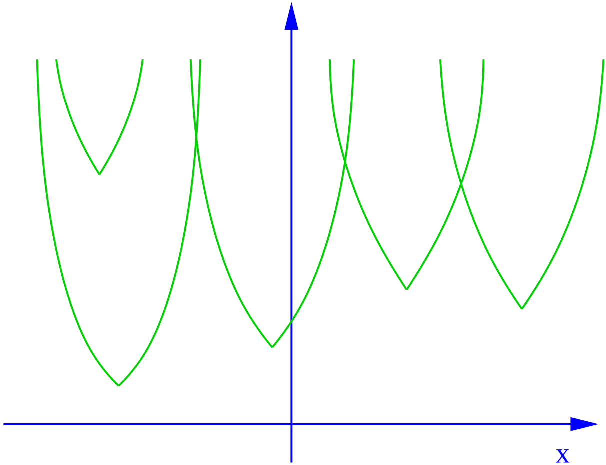

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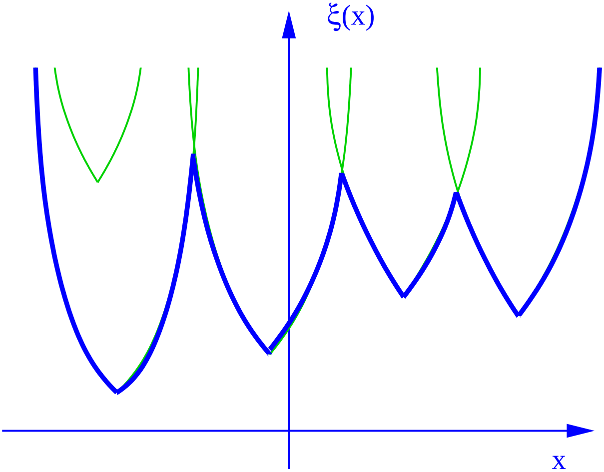

- Assumptions: For all germ $g = (x_g, t_g) \in \mathbb{R}^d \times \mathbb{R}^+$, we assume that

$$\forall t \geq t_g, \qquad C_g(t) = x_g \oplus [V(t) - V(t_g)]K$$

  - K convex compact, $0 \in K^\circ$
  - V absolutely continuous function, $V(t) = \int_0^t v(s)ds$ with speed $0 < v \leq M$

- Consequences: If $t = A_g(x)$, then :

$$[V(t) - V(t_g)]p_{x-x_g,K} = |x - x_g|$$

$$A_g(x) = V^{-1}\left[\frac{|x-x_g|}{p_{x-x_g,K}} + V(t_g)\right]$$

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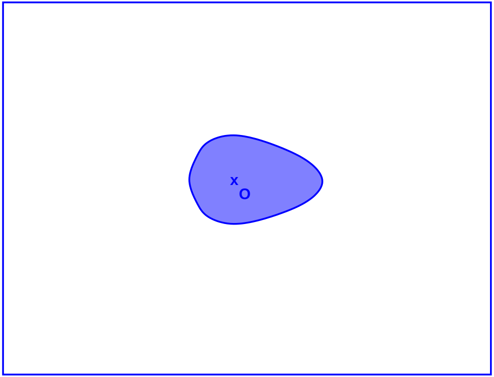

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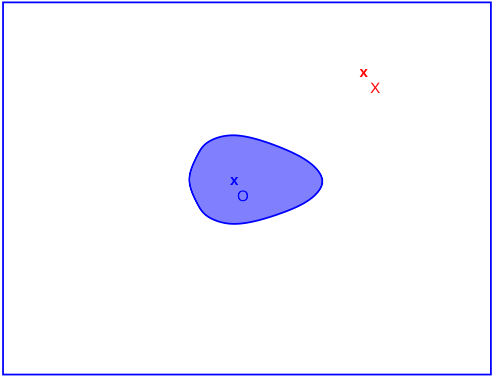

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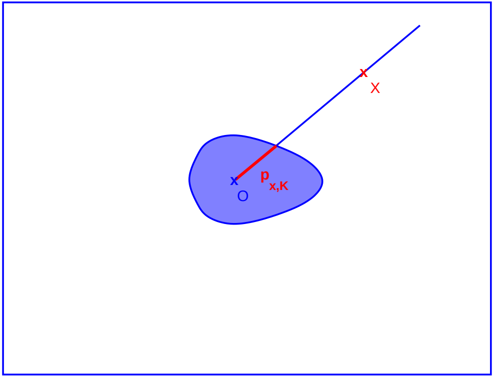

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Theorem 1

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

Sketch of the proof: For all $t > 0$, we introduce the *stationary random field* ξ^t defined by

$$\xi^t(x) = t \wedge \xi(x)$$

1. If, for all $t > 0$, ξ^t is mixing, then ξ is mixing
2. ξ^t is $m(t)$-dependent with $m(t) = 2\,d(t)$ where

$$d(t) = \mathrm{diam}\ C_{(0,0)}(t)$$

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β-mixing coefficients

For two disjoints subsets T_1 and T_2 of $\mathbb{R}^d$, the *absolute regularity coefficient* is:

$$\beta(T_1, T_2) = \|\mathcal{P}_{T_1 \cup T_2} - \mathcal{P}_{T_1} \times \mathcal{P}_{T_2}\|_{var}$$

where $\mathcal{P}_T$ is the distribution of the restriction $\xi_{|T} = (\xi(x))_{x \in T}$.

1. As ξ is stationary, it is sufficient to know $\beta(T_1, T_2)$ up to translations on T_1 and T_2
2. When $d \geq 2$, we consider sets separated in the sense of *Bulinskii (1987)*

β-mixing coefficients

For two disjoints subsets T_1 and T_2 of $\mathbb{R}^d$, the *strong mixing coefficient* is:

$$\alpha(T_1, T_2) = \sup_{A \in \mathcal{F}_{T_1}, B \in \mathcal{F}_{T_2}} |\mathbb{P}(A \cap B) - \mathbb{P}(A)\mathbb{P}(B)|$$

where $\mathcal{F}_{T_i} = \sigma\{\xi(x), x \in T_i\}$ for $i = 1, 2$. Hence, $\alpha(T_1, T_2) \leq \beta(T_1, T_2)$

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Dimension 1

Causal cone

For all $t > 0$, the so-called *causal cone* $K_t = \{g \in \mathbb{R}^+ \times \mathbb{R} \,|\, A_g(0) \leq t\}$ consists of all possible germs that can capture the origin before time t. The measure $\Lambda(K_t)$ is denoted by $\mathcal{G}(t)$.

Theorem 2

If $d = 1$, for two intervals $T_1 = (-\infty, 0]$ and $T_2 = [r, +\infty)$, the coefficient $\beta(T_1, T_2)$ is denoted by $\beta(r)$ and satisfies:

$$\beta(r) \leq C_1 \, \mathrm{e}^{-\mathcal{G}(C_2 \, r)}$$

where $C_1 = 8$ and $C_2 = \frac{1}{2\,M}$

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Dimension 1

Sketch of the proof:

Lemme 1

Let $(\eta(x))_{x\in\mathbb{R}}$ be a random process and T_1 and T_2 two disjoints subsets of $\mathbb{R}$. If there exists two *independent processes* $(\eta_1(x))_{x\in\mathbb{R}}$, $(\eta_2(x))_{x\in\mathbb{R}}$ and two positive constants δ_1, δ_2 such that

$$\mathbb{P}\{\eta(x) = \eta_i(x),\ \ \forall x \in T_i\} \geq 1 - \delta_i \text{ for } i = 1, 2,$$

then

$$\beta(T_1, T_2) \leq 4\,(\delta_1 + \delta_2).$$

Dimension 1

Introduce, for all $T \subset \mathbb{R}$, $\xi_T(x) = \inf_{\substack{g \in \mathcal{N} \\ x_g \in T}} A_g(x)$.

Lemme 2

$$\forall R > 0, \quad \mathbb{P}\{\xi(x) = \xi_{(-\infty,R]}(x), \forall x \leq 0\} \geq 1 - \mathrm{e}^{-\mathcal{G}(R)}$$

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$$\forall R > 0, \quad \mathbb{P}\{\xi(x) = \xi_{[R,+\infty)}(x), \forall x \geq 2\,R\} \geq 1 - \mathrm{e}^{-\mathcal{G}(R)}$$

$$\mathbb{P}\{\xi(0) \leq R\} = \mathbb{P}\{\mathcal{N} \cap K_R \neq \emptyset\} = 1 - \mathrm{e}^{-\mathcal{G}(R)}$$

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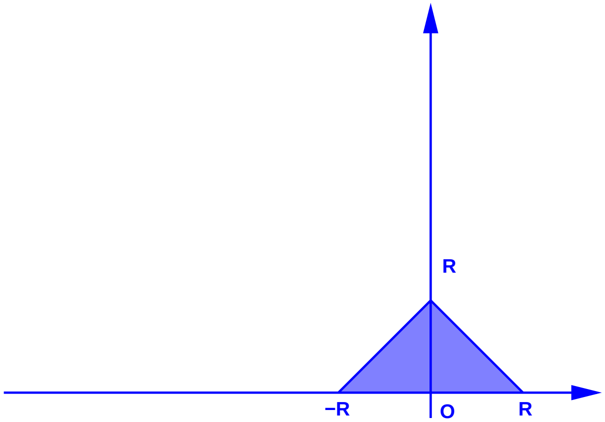

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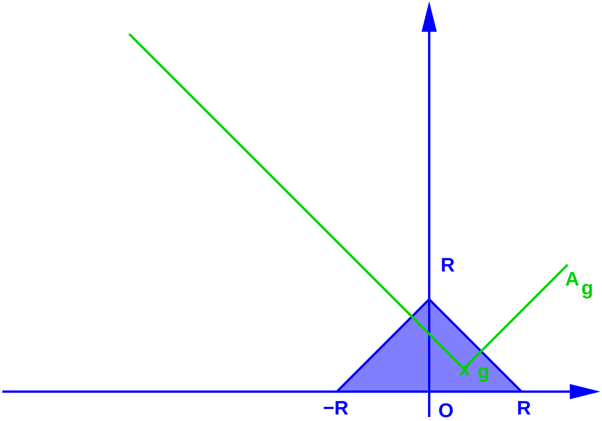

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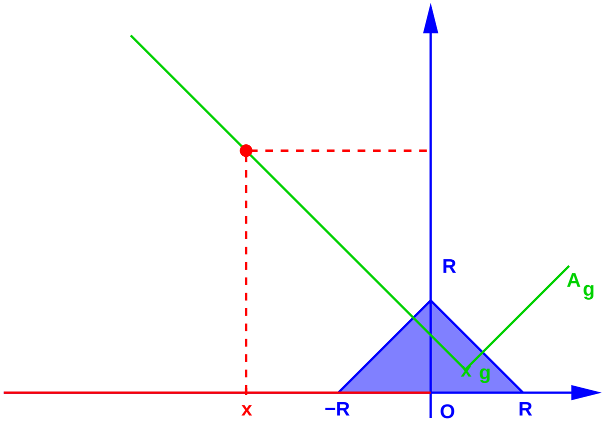

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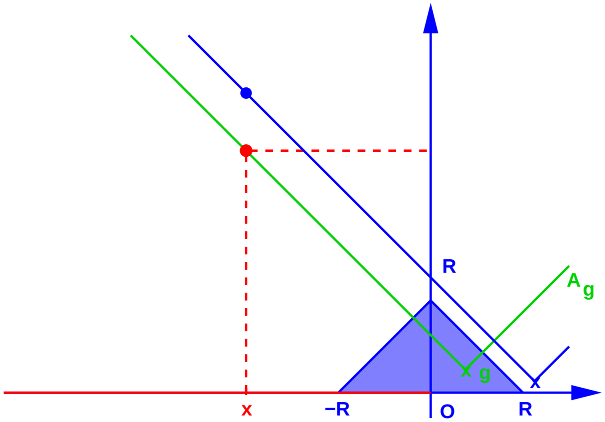

Dimension $d \geq 2$

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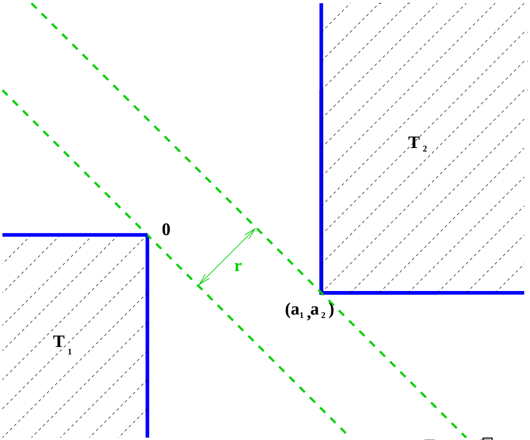
Crystals shape

The crystals shape are defined by the convex compact K:

- D_K is the diameter of the smallest ball centered at zero and containing K
- d_K is the diameter of the greatest ball centered at zero and contained in K
- $A = \frac{D_K}{d_K}$

Dimension $d \geq 2$

Let $T_1 = \prod_{i=1}^{d}(-\infty, 0]$ and $T_2 = \prod_{i=1}^{d}[a_i, +\infty)$ be two quadrants (Q) separated by a r-width band with $r = \frac{\sum_{i=1}^{d} a_i}{\sqrt{d}} > 0$



Dimension $d \geq 2$

Theorem 3

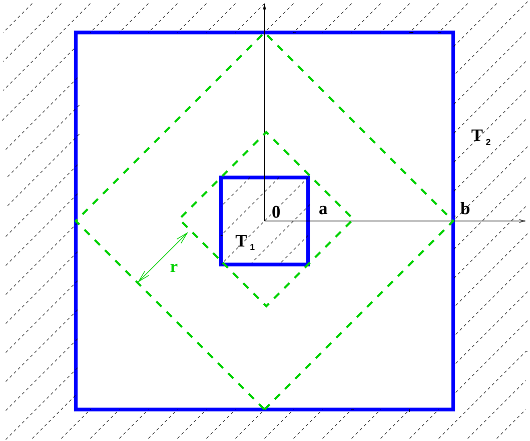
If $d \geq 2$, for two quadrants (Q) $T_1 = \prod_{i=1}^{d}(-\infty, 0]$ and $T_2 = \prod_{i=1}^{d}[a_i, +\infty)$, the coefficient $\beta(T_1, T_2)$ is denoted by $\beta_Q(a, r)$ where a stands for $(a_1, \dots, a_d)$. If $\beta_Q(r) = \sup_{a \in \mathbb{R}^d} \beta_Q(a, r)$, then

$$\beta_Q(r) \leq C_1 \sum_{k=1}^{\infty} k^{d-1} \mathrm{e}^{-\mathcal{G}(C_2(d)\, r\, k)}$$

where $C_1 = 8$ and $C_2(d) = \frac{1}{d\, H^2}$ with $H = 2\,(A + M)$.

Dimension $d \geq 2$

Let $T_1 = [-a, a]^d$ and $T_2 = ([-b, b]^d)^c$ be two enclosed domains (ED) separated by a r-width polygonal band with $r = \frac{(b-2a)\sqrt{d}}{2} > 0$



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$$\beta_Q(r) \leq C_1(d) \sum_{k=1}^{\infty} k^{d-1} \mathrm{e}^{-\mathcal{G}(C_2(d)\, r\, k)}$$

where $C_1(d) = 4(1 + d\, 2^d)$ and $C_2 = \frac{1}{d\, H^2}$ with $H = 2\,(A + M)$.

Intensity measure parameters estimation

The *intensity measure* of the Poisson point process is:

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

Two cases:

1. The measure m is absolutely continuous and $m(dt) = a\, t^{b-1} dt$ with $a, b > 0$
2. The measure m is discrete and $m = \sum_{i=1}^{q} p_i\, \delta_{a_i}$ with $\sum_{i=1}^{q} p_i = 1$, $p_i > 0$ for all $i = 1 \ldots q$ and $0 < a_1 < \cdots < a_q$

We assume that $v = 1$ and $K = B(0, 1)$

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

Two cases:

1. The measure m is absolutely continuous and $m(dt) = a\, t^{b-1} dt$ with $a, b > 0 \Rightarrow \quad a, b$
2. The measure m is discrete and $m = \sum_{i=1}^{q} p_i\, \delta_{a_i}$ with $\sum_{i=1}^{q} p_i = 1$, $p_i > 0$ for all $i = 1 \ldots q$ and $0 < a_1 < \cdots < a_q \Rightarrow \quad p_i, \quad i = 1 \ldots q$

We assume that $v = 1$ and $K = B(0, 1)$

$$\mathcal{F}(t) = \mathbb{P}\{\xi(0) \leq t\}$$
$$= 1 - \mathrm{e}^{-\Lambda(K_t)}$$
$$= 1 - \mathrm{e}^{-\mathcal{G}(t)} \Rightarrow \mathcal{G}(t) = -\log(1 - \mathcal{F}(t))$$

Proposition 1

$$\hat{\mathcal{F}}_n(t) := \frac{1}{n^d} \int_{[0,n]^d} \mathbb{1}_{[0,t]}(\xi(x)) \, \lambda^d(dx)$$

$$\hat{\mathcal{G}}_n(t) := -\log(1 - \hat{\mathcal{F}}_n(t))$$

are strongly consistant estimtors for $\mathcal{F}(t)$ and $\mathcal{G}(t)$:

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Let $(\eta(x))_{x\in\mathbb{R}^d}$ be a stationary random field:

- $\mathbb{E}(\eta(x)) = \mu$
- $R(u) = \mathrm{Cov}(\eta(0), \eta(u))$
- $S_n = \int_{[0,n]^d} (\eta(x) - \mu)\, dx$

We are interested in the asymptotic behaviour of $\frac{S_n}{\sigma n^{\frac{d}{2}}}$ under α-mixing conditions:

- when $d = 1$:

$$\alpha(\rho) = \sup_{A\in\mathcal{F}_{(-\infty,0]},B\in\mathcal{F}_{[\rho,+\infty)}} |\mathbb{P}(A\cup B) - \mathbb{P}(A)\mathbb{P}(B)|$$

- when $d \geq 2$:

$$\alpha_{ED}(\rho) = \sup_{a>0} \alpha_{ED}(a, \rho)$$

Theorem 5

If for some $\delta > 0$,

$$\|\eta(x)\|_{2+\delta} < \infty \tag{1}$$

and

$$\int_0^\infty \rho^{d-1}\,\alpha(\rho)^{\frac{\delta}{2+\delta}}\,d\rho < \infty \tag{2}$$

then $\int_{\mathbb{R}^d} |R(u)|\,du < \infty$. Moreover, if $\sigma^2 = \int_{\mathbb{R}^d} R(u)\,du > 0$, then

$$\frac{S_n}{\sigma n^{\frac{d}{2}}} \xrightarrow[n\to\infty]{\mathcal{D}} \mathcal{N}(0,1).$$

Analogue of Bolthausen's theorem (1982) for continuous-parameter random fields

Theorem 5

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Analogue of Bolthausen's theorem (1982) for continuous-parameter random fields

Corollary 1

Let $(\xi(x))_{x\in\mathbb{R}^d}$ be a stationary random field *satisfying the α-mixing condition*. For all $t \in \mathbb{R}^+$, write

$$\eta_t(x) = \mathbb{1}_{\{\xi(x)\leq t\}} \quad \forall x \in \mathbb{R}^d.$$

Let h be fixed in $\mathbb{N}^*$. If, for $(t_1, \dots, t_h)' \in (\mathbb{R}^+)^d$, the matrix $\Gamma = (\gamma_{i,j})_{i,j=1\dots h}$ which (i,j)-th entry equals

$$\gamma_{i,j} = \int_{\mathbb{R}^d} \mathrm{Cov}\left(\eta_{t_i}(0), \eta_{t_j}(x)\right)\, dx$$

is *positive-definite*, then,

$$n^{\frac{d}{2}}\left((\hat{\mathcal{F}}_n(t_1), \dots, \hat{\mathcal{F}}_n(t_h))' - (\mathcal{F}(t_1), \dots, \mathcal{F}(t_h))'\right) \xrightarrow[n\to\infty]{\mathcal{D}} \mathcal{N}(0, \Gamma).$$

Corollary 2

If $(\xi(x))_{x\in\mathbb{R}^d}$ is a stationary random field *satisfying the α-mixing condition* and the matrix Γ of Corollary 1 is *positive definite*, then

$$n^{\frac{d}{2}}\left((\hat{\mathcal{G}}_n(t_1),\ldots,\hat{\mathcal{G}}_n(t_h))' - (\mathcal{G}(t_1),\ldots,\mathcal{G}(t_h))'\right) \xrightarrow[n\to\infty]{\mathcal{D}} \mathcal{N}(0,V)$$

where the (i,j)-th entry of the covariance matrix $V=(v_{i,j})_{i,j=1\ldots h}$ equals

$$e^{\mathcal{G}(t_i)}\, e^{\mathcal{G}(t_j)}\, \gamma_{i,j}.$$

$m(dt) = a\,t^{b-1}dt$

$$\begin{aligned} \mathcal{G}(t) &= \Lambda(K_t) \\ &= \int_0^t \lambda^d(B(0,t-s))a\,s^{b-1}ds \\ &= c_d\,a\,t^{d+b}\,I_d(b), \end{aligned}$$

where

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

and

$$I_d(b) = \frac{d!}{b\,(b+1)\ldots(b+d)}.$$

For $t = t_1$ and $t = t_2$, we obtain the following system:

$$\begin{cases} b = \dfrac{\log\left(\frac{\mathcal{G}(t_1)}{\mathcal{G}(t_2)}\right)}{\log t_1 - \log t_2} - d \\[2ex] a = \dfrac{\mathcal{G}(t_1)}{c_d \, l_d(b) \, t_1^{d+b}} \end{cases}$$

We introduce the continuous functions

$$g(x_1, x_2) = \frac{\log(\frac{x_1}{x_2})}{\log t_1 - \log t_2} - d$$

and

$$f(x_1, x_2) = \frac{x_1}{c_d \, l_d(g(x_1, x_2)) \, t_1^{d+g(x_1,x_2)}}$$

The system can be summerized under the following form:

$$\begin{cases} a = f(\mathcal{G}(t_1), \mathcal{G}(t_2)) \\ b = g(\mathcal{G}(t_1), \mathcal{G}(t_2)) \end{cases}$$

Proposition 2

The following statistics are strongly consistent estimators for parameters a and b:

$$\begin{aligned} \hat{b}_n &:= g(\hat{\mathcal{G}}_n(t_1), \hat{\mathcal{G}}_n(t_2)) \xrightarrow[n\to\infty]{p.s.} b \\ \hat{a}_n &:= f(\hat{\mathcal{G}}_n(t_1), \hat{\mathcal{G}}_n(t_2)) \xrightarrow[n\to\infty]{p.s.} a. \end{aligned}$$

- When $d = 1$, we get that

$$\alpha(r) \leq \beta(r) \leq C_1\, e^{-\gamma\, r^{1+b}}$$

with $\gamma = -c_d\, a\, C_2^{1+b}\, I_d(b)$.

$$\Rightarrow \int_0^{\infty} \alpha(r)\, dr < \infty$$

- When $d \geq 2$, we obtain that

$$\alpha_{ED}(r) \leq \beta_{ED}(r) \leq C_1(d) \left(\sum_{k=1}^{\infty} k^{d-1} e^{-\gamma(d)\, r^{d+b}(k^{d+b}-1)} \right) e^{-\gamma(d)\, r^{d+b}}$$

with $\gamma(d) = c_d\, a\, I_d(b) C_2(d)^{d+b}$ and for $A > 0$

$$\sup_{r \geq A} \sum_{k=1}^{\infty} k^{d-1} e^{-\gamma(d)\, r^{d+b}(k^{d+b}-1)} < \infty.$$

$$\Rightarrow \int_0^{\infty} r^{d-1} \alpha_{ED}(r)\, dr < \infty$$

- When $d = 1$, we get that

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Theorem 6

Assume, for $h = 2$, that the matrix Γ of Corollary 1 is *positive definite*. Then,

$$n^{\frac{d}{2}} \left((\hat{a}_n, \hat{b}_n) - (a, b) \right) \xrightarrow[n\to\infty]{\mathcal{D}} \mathcal{N}(0, MVM')$$

where V is the matrix defined in Corollary 2 and $M = (m_{i,j})_{i,j=1,2}$ with for $j = 1, 2$,

$$\begin{aligned} m_{1,j} &= \frac{\delta f}{\delta x_j}(\mathcal{G}(t_1), \mathcal{G}(t_2)) \\ m_{2,j} &= \frac{\delta g}{\delta x_j}(\mathcal{G}(t_1), \mathcal{G}(t_2)) \end{aligned}$$

$m = \sum_{i=1}^{q} p_i \, \delta_{a_i}$

$$\begin{aligned} \mathcal{G}(t) &= \Lambda(K_t) \\ &= c_d \sum_{i=1}^{q} p_i \, (t - a_i)^d 1_{\{a_i \leq t\}}. \end{aligned}$$

where

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

For $t = a_i$ with $i = 2 \dots q$, we obtain the following equations:

$$\mathcal{G}(a_i) = c_d \sum_{j=1}^{i-1} p_j \, (a_i - a_j)^d \qquad \forall i = 2 \dots q.$$

Equivalently, we have that

$$\begin{cases} p_1 & = & \dfrac{1}{(a_2 - a_1)^d} \dfrac{\mathcal{G}(a_2)}{c_d} \\ p_i & = & \dfrac{1}{(a_{i+1} - a_i)^d} \left(\dfrac{\mathcal{G}(a_{i+1})}{c_d} - \displaystyle\sum_{j=1}^{i-1} p_j (a_{i+1} - a_j)^d \right) \quad \forall i = 2 \dots q-1 \end{cases}$$

Introducing the following functions,

$$f_1(x_2,\dots,x_q) = \frac{1}{(a_2-a_1)^d}\frac{x_2}{c_d}$$

$$f_i(x_2,\dots,x_q) = \frac{1}{(a_{i+1}-a_i)^d}\left(\frac{x_{i+1}}{c_d} - \sum_{j=1}^{i-1} f_j(x_2,\dots,x_q)(a_{i+1}-a_j)^d\right)$$

$\forall i = 2\dots q-1.$

The previous equations can be rewritten as follows

$$p_i = f_i(\mathcal{G}(a_2),\dots,\mathcal{G}(a_q)) \;\; \forall i = 1\dots q-1.$$

Proposition 3

The following statistics are strongly consistent estimators for parameters p_i :

$$\hat{p}_{i,n} := f_i(\hat{\mathcal{G}}_n(a_2), \ldots, \hat{\mathcal{G}}_n(a_q)) \xrightarrow[n \to \infty]{p.s.} p_i \quad \forall i = 1 \ldots q-1.$$

Moreover,

$$\hat{p}_{q,n} := 1 - \sum_{j=1}^{q-1} \hat{p}_{i,n} \xrightarrow[n \to \infty]{p.s.} p_q.$$

We have that

$$\mathcal{G}(t) = c_d \sum_{i=1}^{q} p_i \, (t - a_i)^d \ \ \forall t > a_q.$$

As a consequence,

$$\mathcal{G}(t) \sim_\infty c_d \, t^d$$

For $d \geq 1$ and r sufficiently large, we get that

$$\beta(r) \leq C \, e^{-\gamma \, r^d},$$

where C and γ are some positive constants.

$$\Rightarrow \int_0^\infty r^{d-1} \alpha(r) \, dr < \infty$$

Theorem 7

Assume, when $h = q - 1$, $t_i = a_{i+1}$ for all $i = 1 \dots q-1$, that the matrix Γ of Corollary 1 is *positive definite*. Then,

$$n^{\frac{d}{2}} \left((\hat{p}_{1,n}, \dots, \hat{p}_{q-1,n})' - (p_1, \dots, p_{q-1})' \right) \xrightarrow[n \to \infty]{\mathcal{D}} \mathcal{N}(0, MVM')$$

where V is the matrix defined in Corollary 2 and M is the matrix which (i,j)-th entry equals

$$m_{i,j} = \frac{\delta f_i}{\delta x_{j+1}} (\mathcal{G}(a_2), \dots, \mathcal{G}(a_q))$$

Example

We assume that:

1. $d = 1$
2. $m = p_1\delta_{a_1} + p_2\delta_{a_2}$ with $0 < a_1 < a_2 = a_1 + 1$

We obtain that:

1. $(\hat{p}_{1,n}, \hat{p}_{2,n}) = (\frac{\hat{\mathcal{G}}_n(a_2)}{2}, 1 - \hat{p}_{1,n}) \xrightarrow[n\to\infty]{p.s.} (p_1, p_2)$.
2. $$\begin{aligned} \sigma^2 &= \int_{\mathbb{R}} \mathrm{Cov}\left(\mathbb{1}_{\{\xi(0)\le a_2\}}, \mathbb{1}_{\{\xi(x)\le a_2\}}\right) dx \\ &= e^{-4\,p_1} \int_0^2 e^{-p_1\,(1-\frac{x}{2})} - 1\, dx = e^{-4\,p_1} f(p_1) > 0 \end{aligned}$$

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3. $$\sqrt{n}(\hat{p}_{1,n} - p_1) \xrightarrow[n\to\infty]{\mathcal{D}} \mathcal{N}(0, (e^{4\,p_1}/4)\,\sigma^2).$$

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3. $$\sqrt{n}(2\,(\hat{p}_{1,n} - p_1)/f(\hat{p}_{1,n})) \xrightarrow[n\to\infty]{\mathcal{D}} \mathcal{N}(0,1).$$