

# Phase separation and oscillations in cluster growth models

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- Introduction and motivation on growth mechanisms
- Longtime behaviour under detailed balance
- Self-similar behavior without detailed balance
- Oscillatory behavior under suitable forcing

# Introduction and Motivation

# Models of nucleation and condensation

## Nucleation of oversaturated vapor:

- Only monomers move
- no collisions between clusters
- clusters grow/shrink by one monomer

## Applications:

- polymerization
- cloud and galaxy formation mechanism

[Smoluchowski 1916, Becker–Döring 1935]

[Tanaka et al J. Chem. Phys. 2011]

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[Smoluchowski 1916, Becker–Döring 1935]

## Clustering of granular gases:

- Single beads hop to neighboring cells
- hopping rate depends on cell filling
- phases: frozen, clustering, gaseous

## Applications:

- migration, population dynamics
- wealth exchange

[Spitzer 1970 zero-range process]

[S Dorbolo et al 2011 Eur. J. Phys. 32]

**Goal:** Dynamic of cluster population.

# Longtime behavior of the exchange-driven growth model

[S. J Nonlinear Sci '19]

# Exchange driven growth model (EDG)

Previous models evolve size distribution  $(X_k)_{k \geq 0}$  in a population of clusters as

$$X_{k-1} + X_l \xrightleftharpoons[K(k,l-1)]{K(l,k-1)} X_k + X_{l-1}, \quad \text{for } k, l \geq 1.$$

- $K(l, k - 1)$  **rate kernel**: jump of monomer from  $l$  to  $k - 1$  cluster

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- $K(l, k-1)$  **rate kernel**: jump of monomer from  $l$  to  $k-1$  cluster
- rate equation is a **countable** set of **nonlinear coupled** ODEs:

$$\begin{aligned} \dot{c}_k = & \sum_{l \geq 1} K(l, k-1) c_l c_{k-1} - \sum_{l \geq 1} K(k, l-1) c_k c_{l-1} \\ & - \sum_{l \geq 1} K(l, k) c_l c_k + \sum_{l \geq 1} K(k+1, l-1) c_{k+1} c_{l-1}. \end{aligned} \tag{EDG}$$

- **two** conservation laws:

$$1 = \sum_{k \geq 0} c_k \quad \text{and} \quad \varrho = \sum_{k \geq 1} k c_k$$



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## Theorem (Well posedness [S. J Nonlinear Sci '19])

If the kernel  $K$  has at most linear growth  $K(k, l) \leq C k l$ , then the solution to (EDG) is a semigroup on  $\mathcal{P}^\varrho = \{c \in \ell^1(\mathbb{N}_0) : c_k \geq 0, \sum_{k \geq 0} c_k = 1, \sum_{k \geq 1} k c_k = \varrho\}$ .

## Equilibria - longtime behavior - phase separation

If the kernel is **curl-free**

$$\begin{array}{ccc}
 X_{k-1} + X_l + X_0 & \xrightleftharpoons[\overleftarrow{K(k,l-1)}]{\overrightarrow{K(l,k-1)}} & X_k + X_{l-1} + X_0 \\
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 \iff
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then there exists a **chemical potential** and (formal) **equilibria**

$$Q_0 = 1, \quad Q_l = \prod_{k=1}^l \frac{K(1, k-1)}{K(k, 0)} \quad \text{and} \quad \omega_l(\phi) = \frac{\phi^l Q_l}{Z(\phi)} \quad \text{with} \quad Z(\phi) = \sum_{l \geq 0} \phi^l Q_l$$

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These equilibria have a **critical mass density**  $\varrho_c \in [0, \infty]$

$$\varrho_c = \limsup_{\phi \uparrow \phi_c} \sum_{l \geq 1} l \omega_l(\phi) \quad \text{with} \quad \phi_c = \lim_{k \rightarrow \infty} \frac{K(k, 0)}{K(1, k-1)} \in (0, \infty].$$

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$\Rightarrow$  **Free energy functional** (gradient flow, Lyapunov function)

$$\mathcal{F}[c] = \sum_{k \geq 0} c_k \log \frac{c_k}{Q_k} = \sum_{k \geq 0} c_k \log c_k + \sum_{k \geq 0} c_k \log \frac{1}{Q_k}.$$

# Longtime behavior and phase separation

## Theorem [S. J Nonlinear Sci '19]

Let  $K$  be curl-free, sublinear, sufficiently regular with  $\rho_c \in (0, \infty)$ .

Then for any  $\varrho \in (0, \infty)$  and any  $c(0) \in \mathcal{P}^\varrho$ , the solution  $c$  of (EDG) satisfies

1. If  $\varrho \leq \varrho_c$ : Then  $\mathcal{F}[c(t)] \rightarrow \mathcal{F}[\omega^\varrho]$  and  $\sum_{l \geq 0} (l+1) |c_l(t) - \omega_l^\varrho| \rightarrow 0$  as  $t \rightarrow \infty$ .
2. If  $\varrho > \varrho_c$ : Then  $\mathcal{F}[c(t)] \rightarrow \mathcal{F}[\omega^{\varrho_c}] + (\varrho - \varrho_c) \log \phi_c$  and  $c_l(t) \rightarrow \omega_l^{\varrho_c}$  for all  $l \geq 0$  as  $t \rightarrow \infty$ .  
In particular no  $\ell^1$ -convergence, since **excess mass**  $\varrho - \varrho_c$  vanishes!

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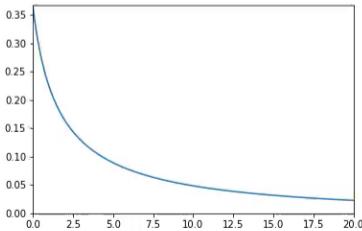
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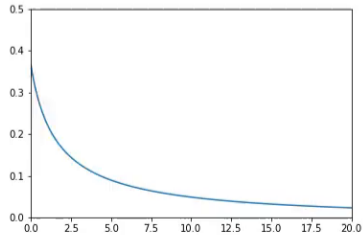
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1



2



# Self-similar behavior of the exchange-driven growth model

[Eichenberg-S. Commun. PDEs '21]



## Special class of kernels [Ben-Naim, Krapivsky '03]



**Product kernel:** for  $\lambda \in [0, 2)$

$$K(k, l) = K_\lambda(k, l) = a_\lambda(k)a_\lambda(l)$$

$$\text{with } a_\lambda(k) = \begin{cases} k^\lambda, & \lambda > 0, \\ 1 - \delta_{k,0}, & \lambda = 0, \end{cases}$$

- $K_\lambda(k, 0) = 0$  for all  $k \geq 1$   
 $\Rightarrow$  no formation of new clusters
- Homogeneity and symmetry  
simplify (EDG) through moments

$$M_\kappa[c] = \sum_{l \geq 1} l^\kappa c_l$$

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$$\begin{cases} \dot{c}_0 = M_\lambda[c] c_1, \\ \dot{c}_1 = M_\lambda[c] (-2c_1 + 2^\lambda c_2), \\ \dot{c}_k = M_\lambda[c] ((k-1)^\lambda c_{k-1} - 2k^\lambda c_k + (k+1)^\lambda c_{k+1}). \end{cases} \quad \text{EDG}_\lambda$$

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$$\ell(t) = \frac{1}{1 - c_0(t)} \sum_{k=1}^{\infty} k c_k(t) = \frac{\rho}{M_0[c]},$$

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## Theorem (Coarsening rate)

$$\ell(t) \propto \begin{cases} t^\beta, & \text{if } 0 \leq \lambda < 3/2, \\ \exp(Ct), & \text{if } \lambda = 3/2, \\ (t_{\text{gel}} - t)^\beta, & \text{if } 3/2 < \lambda < 2, \end{cases}$$

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- Coarsening exponent

$$\beta = \frac{1}{3 - 2\lambda}.$$

# Self-similar behavior

Behave solutions dynamical self-similar?

$$c_k(t) \propto \rho s(t)^{-2} g_\lambda(s(t)^{-1}k) \quad \text{for } t \gg 1?$$

for the explicit profile

$$g_\lambda(x) = \frac{1}{Z_\lambda} \frac{x^{1-\lambda}}{2-\lambda} \exp\left(-\frac{x^{2-\lambda}}{(2-\lambda)^2}\right),$$

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Measure-valued formulation

$$\mu_c(t) = s(t) \sum_{k \geq 1} c_k(t) \delta_{s(t)^{-1} k}.$$

Number of clusters

$$M_0[c] = 1 - c_0 = s^{-1}(t) \int_0^\infty d\mu_c.$$

Total mass density

$$M_1[c] = \int_0^\infty d\mu_c.$$

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## Theorem (Self-similar behavior)

1. For  $0 \leq \lambda < 3/2$  there exists  $C = C(\lambda, \rho) > 0$  and

$$s(t) = Ct^\beta \quad \text{with } \beta = (3 - 2\lambda)^{-1},$$

such that every global solution  $c$  to  $(EDG_\lambda)$  satisfies

$$\mu_c(t) \rightarrow \rho g_\lambda \quad \text{as } t \rightarrow \infty.$$

2. For  $\lambda = 3/2$  ...

3. For  $3/2 < \lambda < 2$  and  $t_{\text{gel}}$  as before there exists

$C = C(\lambda, \rho) > 0$  and

$$s(t) = C(t_{\text{gel}} - t)^\beta \quad \text{with } \beta = (3 - 2\lambda)^{-1},$$

such that every solution  $c$  to  $(EDG_\lambda)$  existing on  $[0, t_{\text{gel}})$

$$\text{satisfies } \mu_c(t) \rightarrow \rho g_\lambda \quad \text{as } t \rightarrow t_{\text{gel}}.$$

## Measure-valued formulation

$$\mu_c(t) = s(t) \sum_{k \geq 1} c_k(t) \delta_{s(t)^{-1}k}.$$

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# Some ingredients for the proof

## Time-change

$$\tau(t) = \int_0^t M_\lambda[c](s) ds.$$

## Discrete weighted Laplacian:

$$\begin{cases} \partial_\tau u = \Delta_{\mathbb{N}}(a_\lambda u), & k \geq 1, \\ u(\tau, 0) = 0, & \tau \geq 0. \end{cases}$$

**Tail-distribution:**  $U(t, k) = \sum_{l \geq k} u(t, l)$

$$\begin{cases} \partial_t U(k) = \partial^-(a_\lambda \partial^+ U)(k), \\ \partial^+ U(t, 0) = 0. \end{cases}$$

## Proposition (Discrete Nash-inequality)

$$\|U\|_2^2 \lesssim \|U\|_1^{\frac{2(2-\lambda)}{3-\lambda}} E_\lambda(U)^{\frac{1}{3-\lambda}},$$

Dirichlet form:  $E_\lambda(U) = \sum_{k \geq 1} k^\lambda |\partial^+ U(k)|^2$ .

$\Rightarrow$  Nash-continuity, decay, moment bounds, ...

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## Continuous solution:

$$\begin{cases} \partial_t \mathcal{U} = \partial_x(a_\lambda \partial_x \mathcal{U}) = \mathcal{L}_\lambda \mathcal{U}, & (t, x) \in \mathbb{R}_+^2, \\ a_\lambda \partial_x \mathcal{U}|_{x=0} = 0, & t \in \mathbb{R}_+, \\ \mathcal{U}(0, \cdot) = \mathcal{U}_0, & x \in \mathbb{R}_+. \end{cases}$$

## Discrete-to-continuum interpolation:

$$\mathcal{U}_\varepsilon(t, x) = \varepsilon^{-\alpha} U(\varepsilon^{-1}t, \lfloor \varepsilon^{-\alpha} x \rfloor + 1).$$

*parabolic scaling:*  $k \propto t^\alpha$  with  $\alpha = \frac{1}{2-\lambda} \in [\frac{1}{2}, \infty)$



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## Theorem

It holds  $\mathcal{U}_\varepsilon \rightarrow \mathcal{U}$ .

**Proof:** Replacement lemma, compactness

**Note:** For  $t = 1$ , we get

$$\varepsilon^{-\alpha} U(\varepsilon^{-1}, \lfloor \varepsilon^{-\alpha} x \rfloor + 1) \rightarrow \rho \mathcal{G}_\lambda(x).$$

$\Rightarrow$  Obtain long-time behavior by setting  $t = \varepsilon^{-1}$ .

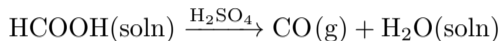
# Oscillations in a Becker–Döring model with injection and depletion

[Niethammer-Pego-S.-Velazquez arXiv:2102.06751]

# From the Morgan reaction to the Bubbleator



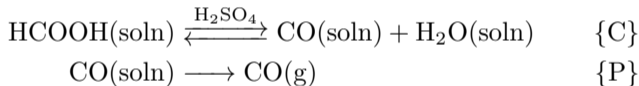
**Morgan reaction**<sup>†</sup> describes the oscillatory release of gas CO during dehydration of formic acid in concentrated sulfuric acid



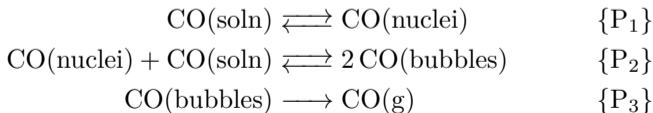
<sup>†</sup>[Morgan, J. S. *J. Chem. Soc., Trans.* 1916, 109, 274-83.]

<sup>b</sup>[Smith, K. W.; Noyes, R. M.; Bowers, P. G. *J. Phys. Chem.*, “Chemical Oscillations and Instabilities”, 1983.]

SNB<sup>b</sup> separated reaction into reversible and irreversible parts



Process {P} is separated into a growth process of CO-bubbles

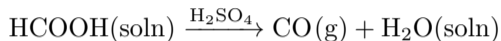


**Goal:** Minimal models for {P<sub>1</sub>-P<sub>3</sub>} showing oscillations.

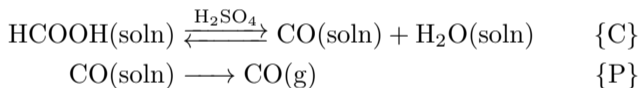
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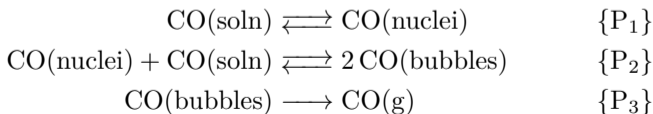
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<sup>†</sup>[Morgan, J. S. *J. Chem. Soc., Trans.* 1916, 109, 274-83.]

<sup>b</sup>[Smith, K. W.; Noyes, R. M.; Bowers, P. G. *J. Phys. Chem.*, "Chemical Oscillations and Instabilities", 1983.]

[Kunz, H.; Johannesmeyer, F.; Oetken, M.: Das „pulsierende Sektglas“, CHEMKON 7 (2000) 30-31]

# Modeling of the Bubbleator

## Ingredients for oscillations in chemical reaction networks:

- open system
- no detailed balance
- feed-back loop (nonlinear)
- multiple stationary points

Bubbleator is a physical-chemical system:<sup>1</sup>

**Chemical:** reaction provides a constant source of microbubbles

**Physical:** larger bubbles form through coagulation of microbubbles

*Oscillations are caused by the growth process of the bubbles.*

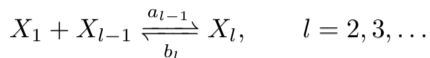
**Goal:** Oscillatory growth model for the statistics of bubble sizes with constant source.

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<sup>1</sup>Bowers, P. G.; Noyes, R. M.: *Chapter 13: Gas Evolution Oscillators*. Wiley, New York (1985) 473-492.

# Becker-Döring equation – derivation

Model [Becker–Döring '35] for **coagulation** and **fragmentation** of clusters consisting of identical monomers (microbubbles)



under the assumption of **conservation of the total mass density**

$$\sum_{l=1}^{\infty} l n_l(t) = \sum_{l=1}^{\infty} l n_l(0) = \rho_0.$$

Let  $J_l$  be the **net-flux** from  $l-1$  to  $l$ -clusters

$$\dot{n}_l(t) = J_{l-1}(t) - J_l(t) \quad l = 2, 3, \dots$$

Mass conservation implies

$$\dot{n}_1(t) = - \sum_{l=1}^{\infty} J_l(t) - J_1(t) =: J_0(t) - J_1(t).$$

The (net) flux is determined from **mass-action-kinetics**

$$J_l(t) = a_l n_1(t) n_l(t) - b_{l+1} n_{l+1}(t), \quad l = 1, 2, \dots$$

# Becker-Döring equation – stationary states and rates



Stationary states are characterized by the detailed balance condition  $J_l = 0$

$$a_l \omega_l \omega_l = b_{l+1} \omega_{l+1} \quad \Rightarrow \quad \omega_l(z) = z^l Q_l \quad \text{with } Q_1 = 1 \text{ and } Q_l = \frac{a_{l-1} \cdots a_1}{b_l \cdots b_2}, l \geq 2.$$

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$\Rightarrow$  Does  $\omega(z)$  have finite mass?

**Assumption:**

$$\text{finite convergence radius: } z_s = \lim_{l \rightarrow \infty} \frac{b_l}{a_l} \in (0, \infty) \quad \text{and critical mass } \rho_s := \sum_{l=1}^{\infty} l \omega_l(z_s) < \infty.$$

**Concrete (physical) rates:** For  $\alpha \in [0, 1)$ ,  $\gamma \in (0, 1)$  and  $z_s, q > 0$

$$a_l := l^\alpha \quad \text{and} \quad b_l := l^\alpha (z_s + q l^{-\gamma}).$$

**Example:** Exponents 3-dim (interface-reaction-limited growth):  $\alpha = \frac{2}{3}, \gamma = \frac{1}{3}$

Stationary state for  $l \gg 1$

$$\omega_l(z) \simeq \exp\left(l \log\left(\frac{z}{z_s}\right) - \frac{q}{1-\gamma} l^{1-\gamma}\right).$$

Define  $z(\rho_0)$  such that  $\sum l \omega_l(z) = \rho_0$  for  $\rho_0 \leq \rho_s$  and set  $z(\rho_0) = z_s$  for  $\rho_0 > \rho_s$ .



# Becker-Döring equation – long-time behavior

Consider **free energy**

$$\mathcal{F}(n) := \mathcal{H}(n|\omega(z)) := \sum_{l=1}^{\infty} \omega_l \eta\left(\frac{n_l}{\omega_l}\right) \quad \text{with} \quad \eta(x) = x \log x - x + 1.$$

Then 
$$\frac{d}{dt} \mathcal{F}(n) = - \sum_{l=1}^{\infty} (a_l n_1 n_l - b_{l+1} n_{l+1}) (\log a_l n_1 n_l - \log b_{l+1} n_{l+1}) \leq 0$$

## Long-time behavior [Ball, Carr, Penrose '89]

- For  $z = z(\rho_0)$  as before holds  $\mathcal{F}(n) \rightarrow 0$  as  $t \rightarrow \infty$ .
- In the case  $\rho_0 > \rho_s$  holds  $n_l(t) \rightarrow \omega_l(z_s)$  for  $l \geq 1$  as  $t \rightarrow \infty$ .  
In particular  $\rho_0 = \sum_l l n_l(t) \not\rightarrow \sum_l l \omega_l(z_s) = \rho_s$ .  
Moreover, the infimum

$$\inf_{n \in \mathcal{M}(\rho_0)} \mathcal{F}(n) = \mathcal{F}(\omega(z_s)) \quad \text{is not attained!}$$

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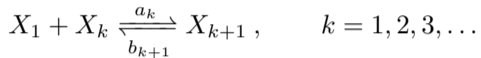
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[Niethammer '03, S. '19] find a nonlocal conservation law (LSW) for the excess mass.

# The Becker-Döring-Bubbleator: Model

BD-bubbleator: source  $S > 0$  and depletion  $r > 0$



## Reaction rate ODE system

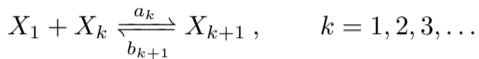
$$\dot{n}_1 = -J_1[n] - \sum_{k=1}^{\infty} J_k[n] + S$$

$$J_k[n] = a_k n_1 n_k - b_{k+1} n_{k+1}, \quad k = 2, 3, \dots$$

$$\dot{n}_k = J_{k-1}[n] - J_k[n] - rk^b n_k, \quad k = 2, 3, \dots$$

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Assumptions and regime:

Physical rates (surface area, Gibbs-Thompson):

$$a_k = k^\alpha \quad \text{and} \quad b_k = a_k \left(1 + \frac{q}{k^\gamma}\right).$$

Stationary distribution ( $S = 0, r = 0$ ):  $n_1 = 1$

$$\omega_l(n_1) \simeq \exp\left(l \log(n_1) - \frac{q}{1-\gamma} l^{1-\gamma}\right).$$

Let  $n_1 = 1 + \varepsilon > 1$ : critical  $k_{\text{crit}}(n_1)$  is

$$a_{k_{\text{crit}}} n_1 - b_{k_{\text{crit}}} = 0 \quad \Leftrightarrow \quad k_{\text{crit}} = \left(\frac{q}{\varepsilon}\right)^{\frac{1}{\gamma}}.$$

Second order expansion: **excess defect**  $u$

$$n_1 = 1 + \varepsilon + \left(\frac{\varepsilon}{q}\right)^{\frac{1}{\gamma}} u = 1 + \varepsilon + \frac{u}{k_{\text{crit}}}.$$

# Constant flux steady states: Nucleation flux

**Recall:** Zero flux steady states are characterized by the **detailed balance condition**  $J_l = 0$

$$a_l \omega_l \omega_l = b_{l+1} \omega_{l+1}$$

$$\Rightarrow \omega_l(z) = z^l Q_l$$

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# Constant flux steady states: Nucleation flux

**Now:** Constant flux steady states  $\{f_k\}_{k \geq 1}$  solve  $J_k[f] = J(\bar{n}_1) \neq 0$

$$a_k \bar{n}_1 f_k(\bar{n}_1) - b_{k+1} f_{k+1}(\bar{n}_1) = J_k[f] = J(\bar{n}_1), \quad l \geq 2,$$

$f_1(\bar{n}_1) = \bar{n}_1$  and such that  $f_k(\bar{n}_1)$  is bounded as  $k \rightarrow \infty$ .

$$\Rightarrow f_k(\bar{n}_1) = J(\bar{n}_1) Q_k \bar{n}_1^k \sum_{l=k}^{\infty} \frac{1}{a_l Q_l \bar{n}_1^{l+1}}$$

where 
$$\frac{1}{J(\bar{n}_1)} = \sum_{l=1}^{\infty} \frac{1}{a_l Q_l \bar{n}_1^{l+1}}.$$

By Laplace asymptotics follows for  $n_1 = 1 + \varepsilon + \left(\frac{\varepsilon}{\gamma}\right)^{\frac{1}{\gamma}} u$

$$J(n_1) \simeq J_{\infty} e^u \quad \text{with } J_{\infty} = \sqrt{\frac{\gamma}{2\pi q^{\frac{1}{\gamma}}}} \varepsilon^{\frac{\gamma+1}{2\gamma}} \exp\left(-\frac{\gamma}{1-\gamma} q^{\frac{1}{\gamma}} \varepsilon^{-\frac{1-\gamma}{\gamma}}\right).$$

and 
$$f_k(\bar{n}_1) \simeq \frac{J(\bar{n}_1)}{\varepsilon a_k} = \frac{J(\bar{n}_1)}{\varepsilon k^{\alpha}} \quad \text{for } k \gg k_{\text{crit}}.$$

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with  $Q_1 = 1$  and  $Q_l = \frac{a_{l-1} \cdots a_1}{b_l \cdots b_2}.$

# Quasistationary approximation

## Transport approximation of large clusters:

For the flux for cluster much larger the critical volume

$k \gg R \gg k_{\text{crit}}$  holds

$$J_k = a_k(n_1 - 1)n_k - \frac{a_k q}{k^\gamma} n_k + b_k n_k - b_{k+1} n_{k+1} \simeq a_k \varepsilon n_k.$$

$\Rightarrow$  transport equation on macroscopic scale: Set  $f(k, t) = n_k(t)$

$$\partial_t f(y, t) + \varepsilon \partial_y (y^\alpha f(y, t)) = -r y^b f(y, t).$$

coupled through boundary layer

$$f(y, t) \simeq J(n_1) \frac{1}{\varepsilon y^\alpha} \simeq J_\infty e^u \frac{1}{\varepsilon y^\alpha} \quad \text{for } k_{\text{crit}} \ll y \ll R.$$

## Exponential monomer excess:

Introduce cut-off  $L \ll 1$  of sub- and supercritical clusters:

$$\frac{\partial_t u}{k_{\text{crit}}} = -(L k_{\text{crit}} + 1) J(n_1) - \varepsilon \int_{L k_{\text{crit}} + 1}^{\infty} f(y, t) y^\alpha dy + S.$$

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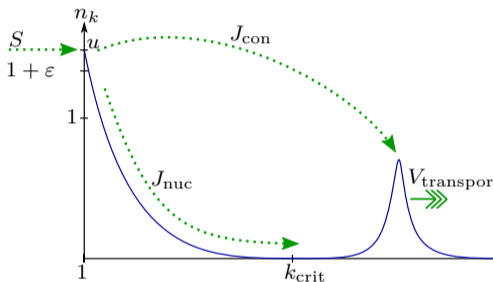
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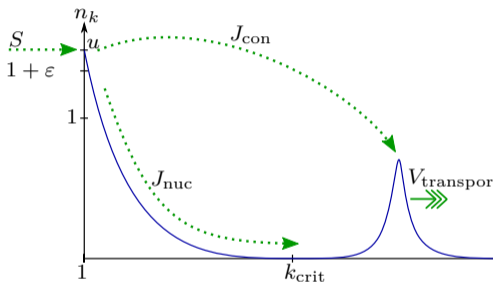
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## Quasistationary approximation:



## Identification of scales:

- $J_{\text{nuc}} \approx J_{\text{con}} \approx 1 \Rightarrow$  fixes time and length-scale
- $S$  such that  $u = O(1) \Rightarrow$  fixes  $S > 0$

# Limit model and Hopf bifurcation

**Model after setting the scales  $(R, T, S)$ :**

Evolution of exponential monomer excess:

$$\partial_\tau u = 1 - \int_0^\infty g(x, \tau) x^\alpha dx$$

Evolution of supercritical cluster density for  $x = R^{-1}y > 0$ :

$$\partial_\tau g(x, \tau) + \partial_x (x^\alpha g(x, \tau)) = -(TR^b r) x^b g(x, \tau)$$

Boundary behavior:  $g(x, \tau) \simeq \frac{e^u}{x^\alpha}$  for  $x \rightarrow 0$

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## Limit model

$$\partial_\tau u = 1 - \int_0^\infty h(z, \tau) z^\nu dz,$$

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**Integration of characteristics:**

$$h(z, \tau) = \exp(u(\tau-z)) \exp\left(-\frac{\theta}{\beta+1} z^{\beta+1}\right)$$

gives in the equation of the monomer excess:

$$\partial_\tau u = 1 - \int_0^\infty e^{u(\tau-z)} \exp\left(-\frac{\theta}{\beta+1} z^{\beta+1}\right) z^\nu dz$$

**Constant stationary solution:** For any  $\theta > 0$

$$1 = e^{u_0} \int_0^\infty \exp\left(-\frac{\theta}{\beta+1} z^{\beta+1}\right) z^\nu dz.$$

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**Starting point for Hopf analysis:**

- Linearization around constant solution
- Find pure imaginary eigenvalues  $\Rightarrow$  critical  $\theta^*$
- Prove **transversality condition**: critical eigenvalues get positive real part for  $\theta < \theta^*$   $\Rightarrow$  second order expansion of  $U$ .
- $\beta$  odd has **infinitely many bifurcations**

# Hopf-Bifurcation of the limit model

**Parameters:**  $\theta = 0.1$ ,  $n = 2$ ,  $\beta = 1$

# The Becker-Döring-Bubbleator: Model II

Consider the truncated Becker-Döring model on  $\{1, 2, \dots, M\}$



with constant source  $S > 0$  of monomers:

$$\dot{n}_1 = -J_1[n] - \sum_{k=1}^{M-1} J_k[n] - J_M^{\text{out}}[n] + S$$

$$J_k[n] = a_k n_1 n_k - b_{k+1} n_{k+1}, \quad \text{for } k = 2, \dots, M-1$$

$$\dot{n}_k = J_{k-1}[n] - J_k[n] \quad \text{for } k = 2, \dots, M-1$$

$$\dot{n}_M = J_{M-1}[n] - J_M^{\text{out}}[n] \quad \text{with } J_M^{\text{out}}[n] = a_M n_1 n_M.$$

Oscillations can be observed for the rates:

- strong dissipative dynamic of small clusters  $k = 1, \dots, K$
- transport/hyperbolic dominated dynamic of large clusters  $k = K+1, \dots, M$

$$a_l = A, \quad l = 1, \dots, K-1 \quad \text{and} \quad a_l = 1, \quad l = K, \dots, M$$

$$b_l = A, \quad l = 2, \dots, K \quad \text{and} \quad b_l = 0, \quad l = K+1, \dots, M$$

# Hopf-Bifurcation in $M = 500, K = 15$



Unstable stationary state  $M = 500, K = 30$

# The Becker-Döring-Bubbleator: Stationary states

$$\begin{aligned} \partial_t n_1 &= -A \left[ (n_1)^2 - n_2 + \sum_{\ell=1}^{K-1} (n_1 n_\ell - n_{\ell+1}) \right] - n_1 \sum_{\ell=K}^M n_\ell + S \\ \partial_t n_\ell &= A [(n_1 n_{\ell-1} - n_\ell) - (n_1 n_\ell - n_{\ell+1})] \quad , \quad 2 \leq \ell \leq K-1 \\ \partial_t n_K &= A (n_1 n_{K-1} - n_K) - n_1 n_K \\ \partial_t n_\ell &= n_1 (n_{\ell-1} - n_\ell) \quad , \quad K+1 \leq \ell \leq M \end{aligned}$$

Constant flux stationary state is explicit given by

$$\begin{aligned} \frac{S}{M+1} &= n_1^{K+1} \frac{1 - n_1}{1 - (1 - \frac{1}{A})n_1 - \frac{n_1^K}{A}} \\ n_l &= n_1^l - \frac{S}{M+1} \frac{1 - n_1^{l-1}}{1 - n_1} \quad , \quad \text{for } l = 2, \dots, K \\ n_l &= n_K \quad , \quad \text{for } l = K+1, \dots, M . \end{aligned}$$

# Quasi-stationary approximation $A \rightarrow \infty$

Small clusters become quasistationary in the limit  $A \rightarrow \infty$

$$n_l = n_1^l \quad \text{for } l = 2, \dots, K$$

Allows to arrive at simplified systems, after deriving equation for  $n_1$

$$\partial_t n_1 = \frac{S - n_1 \Phi - (n_1)^{K+1}}{\sum_{\ell=1}^K \ell^2 (n_1)^{\ell-1}} \quad \text{with } \Phi = \sum_{\ell=K}^M n_\ell$$

$$\partial_t n_\ell = n_1 (n_{\ell-1} - n_\ell), \quad K+1 \leq \ell \leq M$$

$$n_K = (n_1)^K$$

which has a constant flux stationary state given by

$$N_1 = \left( \frac{S}{M - K + 2} \right)^{\frac{1}{K+1}} \quad \text{and} \quad N_l = N_1^K \quad \text{for } l = K, \dots, M.$$

Linearization in the regime  $M \gg 1$  shows a Hopf-bifurcation in the parameter  $K$ .

Eigenvalues for  $M \gg 1$  solve approximately

$$\frac{1}{K} (\alpha z^2 + z) + (1 - e^{-z}) = 0 \quad \text{with} \quad \alpha = \frac{1}{M^2} \sum_{m=0}^{K-1} (K-m)^2 (N_1)^{-m}$$