Dealing with probability of multiple fragmentation in liquid droplet dynamics

A “close–up” view of breakage and scattering kernels

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A very short bibliography!

HISTORICAL PAPERS:

[1] **M. von Smoluchowski**. Versuch einer mathematischen theorie der kogulationskinetic kolloid lösungen. *Z. Phys. Chem.*, 1917

[2] **S. Chandrasekhar**. Stochastic processes in physics and astronomy. *Rev. Modern Phys.*, 1943.

[3] **Z. A. Melzak**. A scalar transport equation. *Trans. Amer. Math. Soc.*, 1957.

INTERESTING PAPERS BY CHEMICAL ENGINEERS:

[4] **K. J. Valentas and R. N. Amundson**. Breakage and coalescence in dispersed phase systems. *I& E C Fundamentals*, 1966.

[5] **C. Tsouris and L. T. Tavlarides**. Breakage and coalescence models for drops in turbulent dispersions. *AIChE Journal*, 1994.

[6] **S. Kumar, R. Kumar, and K. S. Gandhi**. A new model for coalescence efficiency of drops in stirred dispersions. *Chemical Engineering Science*, 1993.

[7] **Sajeev Kumar, R. Kumar, and K. S. Gandhi**. A multi–stage model for drop breakage in stirred vessels. *Chemical Engineering Science*, 1992.

OUR CONTRIBUTION:

[8] **A. Fasano and F. Rosso**. A new model for the dynamics of dispersions in a batch reactor. *Lectures in Applied Mathematics*, 2000.

[9] **A. Fasano and F. Rosso**. Breakage, coalescence and volume scattering in dispersed phase systems. *XXI Southeastern conference on theoretical and applied mechanics*, Developments on Theoretical and Applied Mechanics, 2002.

[10] **A. Fasano and F. Rosso**. Dynamics of droplets in an agitated dispersion with multiple breakage. Part I: formulation of the model and physical consistency. *Math. Meth. Appl. Sc.* (2004, to appear)

[11] **A. Fasano and F. Rosso**. Dynamics of droplets in an agitated dispersion with multiple breakage. Part II: uniqueness and global existence. *Math. Meth. Appl. Sc.* (2004, to appear)

RECENT PAPERS THAT USE OUR MODEL

[12] **C. Walker**. Coalescence and breakage processes. *Mathematical Methods in the Applied Sciences*, 2002.

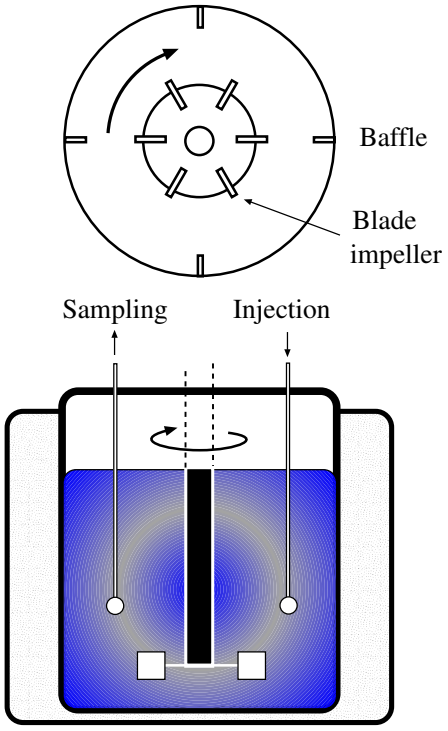
[13] **H. Amann and C. Walker**. Local and Global Strong Solutions to Continuous Coagulation–Fragmentation Equations with Diffusion (2004, to appear)

[14] **P. Laurençot and C. Walker**. Steady State for Coagulation–Fragmentation Equation with Volume Scattering (2004, to appear)

- **Physical situation**: Two immiscible liquids (say WATER and OIL) are mixed in some container. Depending on the hold–up (mutual percentage of the two phases) AGITATION yields a DISPERSION of one liquid into the other.
- **problem**: GIVEN $f_o(v,\mathbf{x})$ — the initial number of droplets of dispersed phase with size in $(v, v+\mathrm{d}v)$ per unit volume of the continuous (*guest*) phase at point $\mathbf{x}$ — FIND $f(v,\mathbf{x},t)$ for t sufficiently large.
- **Industrial interest**: dispersion are rather common in pharmacology, chemistry, food manifacturing, crude oil transportation, paintings, ...!

Typical “apparatus”: a blade impeller reactor

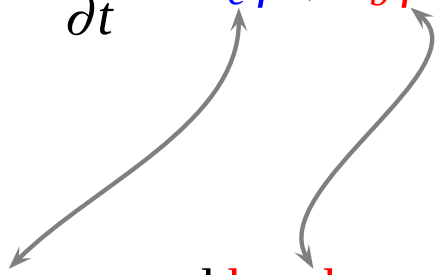
In many interesting cases the very high agitation speed (thousands of R.P.M.) allows to forget diffusion (the system can be considered as SPATIALLY HOMOGENEOUS)



Two guide physical processes: **COLLISIONS.** and **BRAKAGE** (see movies)

standard model (before F. & R.) (without diffusion)

$$\frac{\partial f}{\partial t} = L_c f + L_b f$$



Competing effects: coalescence and breakage.

Is the classical model suitable to fully explain the observed dynamics?

Answer: NO!

No satisfactory explanation for the instability of "large droplets"!

our model (without diffusion)

$$\frac{\partial f}{\partial t} = \phi(t)\,(L_c f + L_b f + L_s f)$$

NEW TERMS: efficiency factor and volume scattering

$\phi(t) = \Psi[\mathscr{N}(t), \mathscr{S}(t)]$ where

$$\mathscr{N}(t) = \int_0^{v_m} f(v,t)\,\mathrm{d}v\,, \qquad \mathscr{S}(t) = \int_0^{v_m} v^{(2/3)} f(v,t)\,\mathrm{d}v\,,$$

State of the art as far as the mathematics is concerned:

Under a “reasonable” set of hypotheses, the evolution equation with the initial condition $f(v,0) = f_o(v)$ has one and only one solution with or without diffusion (Fasano and R. 1998–2004 for the “new” model, many other Authors for the “simplified” model in which $L_s f = 0$ and $\Psi \equiv 1$)

Our current business: what is the very structure of L_b and L_s?

Both are integral operators (linear the former, quadratic the latter) but the structure of their kernels is sistematically left rather “generic” (several reasons for this). However to investigate the kernel structure

- **would help to understand “intimately” the dynamics**
- **would provide useful insights “when doing mathematics” towards the right choice of hypotheses about all terms appearing in the kernels themselves**

Price to pay for this: the mathematics becomes becomes very complex!
Let’s see why.

Structure of the coalescence operator

$$L_c f(v,t) = \underbrace{\int_0^{v/2} \tau_c(w, v-w) f(w,t) f(v-w,t)\, \mathrm{d}w}_{\text{gain}}$$

$$\underbrace{- f(v,t) \int_0^{v_m - v} \tau_c(w,v) f(w,t)\, \mathrm{d}w}_{\text{loss}}$$

Coalescence is always binary: [w] + [v-w] produce the gained drop [v], while [v] disappears because of its merging with [w] to produce [v+w]. Due to the finite size limit v_m, w cannot exceed $v_m - v$.

Structure of the breakage operator

Breakage is described by

$$L_b f(v,t) = \underbrace{\int_v^{v_m} \mathfrak{O}_b^+(s,v) f(s,t)\, \mathrm{d}s}_{\text{gain}} \underbrace{-\mathfrak{O}_b^-(v) f(v,t)}_{\text{loss}}$$

$\mathfrak{O}_b^+(s,v)$ is the gain breakage kernel: **in the current literature nobody distinguishes among the various breakage modes**! This means that, being $\boxed{\text{s}}$ the size of the parent drop and $\boxed{\text{v}}$ that of the observed daughter, $\boxed{\text{s-v}}$ **is the cumulative size of other daughters regardless of how many they are**! Similarly $\mathfrak{O}_b^-(v)$ is the loss breakage kernel: in this form it is not possible to distinguish the contributions to the disappering of v due to different fragmentation modes !

Structure of the scattering operator

$$L_s f(v,t) = \underbrace{\int_{v_m}^{v_m+v} \int_{s-v_m}^{s/2} \mathfrak{O}_s^+(s,v,w)\, f(w,t)\, f(s-w,t)\, \mathrm{d}w\, \mathrm{d}s}_{\text{gain}}$$

$$\underbrace{- f(v,t) \int_{v_m-v}^{v_m} \tau_c(w,v)\, f(w,t)\, \mathrm{d}w}_{\text{loss}}$$

$\mathfrak{O}_s^+(s,v,w)$ is the gain scattering kernel: when s is above v_m it becomes unstable and breaks up in many pieces. One of these is v, the remaining ones amount to w. But $\mathfrak{O}_s^+(s,v,w)$ still hides the contributions to w due to each breakage mode.

Although the L_s term (and its implications) was the main contribution of our research on this topic, only recently we have been able to understand the true structure of the $\mathfrak{O}$–kernels. Without this understanding it does not make too much sense to make hypotheses upon the various kernels only guided by “mathematical necessities”! Let’s see the “intimate” structure of kernels

$$\mathfrak{O}_{\mathrm{b}}^{+}(s,v) = \alpha_2(s)\beta_2(s,v)$$

$$+ \sum_{k=3}^{N} \alpha_k(s) \int_{D_k(s,v)} \beta_k\left(s, u_1, \ldots, u_{k-2}, s - v - U_{k-2}\right) \, \mathrm{d}\sigma_{k-2},$$

$$\mathfrak{O}_{\mathrm{b}}^{-}(v) = \sum_{k=2}^{N} \alpha_k(v),$$

$$\mathfrak{O}_{\mathrm{s}}^{+}(s,v,w) = \tau_c(s-w,w)\left(\lambda_2(s)\beta_2(s,s-v)\right.$$

$$\left. + \sum_{k=3}^{N} \lambda_k(s) \int_{D_k(s,v)} \beta_k\left(s, u_1, \ldots, u_{k-2}, s - v - U_{k-2}\right) \, \mathrm{d}\sigma_{k-2} \right)$$

These kernels are "easy" to read!

- $\alpha_k(v)$ is the **breakage frequency of the i–th mode**
- $\beta_k(s, u_1, \ldots, u_{k-1})$ is the **breakage probability density** of drops with volume $s \in (0, 2v_m)$ to generate by breakage k fragments with prescribed volumes u_j $(j = 1, \ldots, k-1)$ *in increasing order*
- $\tau_c(v, w)$ is the **coalescence kernel**
- $\lambda_k(s)$ is a suitable **weight** (to be chosen conveniently) measuring the chance of the parent droplet $s \in (v_m, 2v_m]$ to break exactly in k pieces within the scattering process $\left(\sum_{k=2}^{N} \lambda_k(s) = 1 \right)$

Notice that α_k and λ_k appears in similar contexts but they have different: meanings: "rates" α_k **include the breakage frequency**, while in the operator L_s the breakage has probability one and all we need to know is the probability of each breakage mode.

MAIN ADVANTAGE: the hypotheses needed to prove existence and uniqueness are placed **directly** on α_k, $\beta_k(s, u_1, \ldots, u_{k-1})$ and λ_k and turns out to be **"extremely readable"** and **"easy to check"**. Moreover **their "form" is suggested by the physics**.

Hypotheses placed **directly** on the whole kernels $\mathfrak{O}_b^+$ and $\mathfrak{O}_s^+$ are usually **very far from being "readable"** or **"easy to check"**. **Their "form" is non suggested by the physics** because of their very "cumulative" nature.

MORE COMPLICATE IS TO DESCRIBE THE INTEGRATION DOMAINS!

Integrals over $D_k(s, v)$ do not appear **if and only if** breakage and volume scattering do not involve modes higher than order two (binary events!). We know that these events alone are almost impossible. Thus **we cannot get rid of higher order terms**!

EASIEST CASE: $N = 2$ (binary events)

$$\beta_2(s,u) = \beta_2(s,s-u), \qquad \beta_2(s,u) = 0, \quad \text{if } s \le u \tag{0.1}$$

Assigned over

$$T_{2,1}(s) = \{u_1 \mid 0 < u_1 \le s - u_1 \le v_m\} = \left(\max\{0, s - v_m\}, \frac{s}{2}\right), \tag{0.2}$$

then **extended by axial simmetry** (with respect to $s/2$) over

$$T_{2,2}(s) = \{u_1 \mid 0 < s - u_1 < u_1 \le v_m\} = \left(\frac{s}{2}, \min\{s, v_m\}\right), \tag{0.3}$$

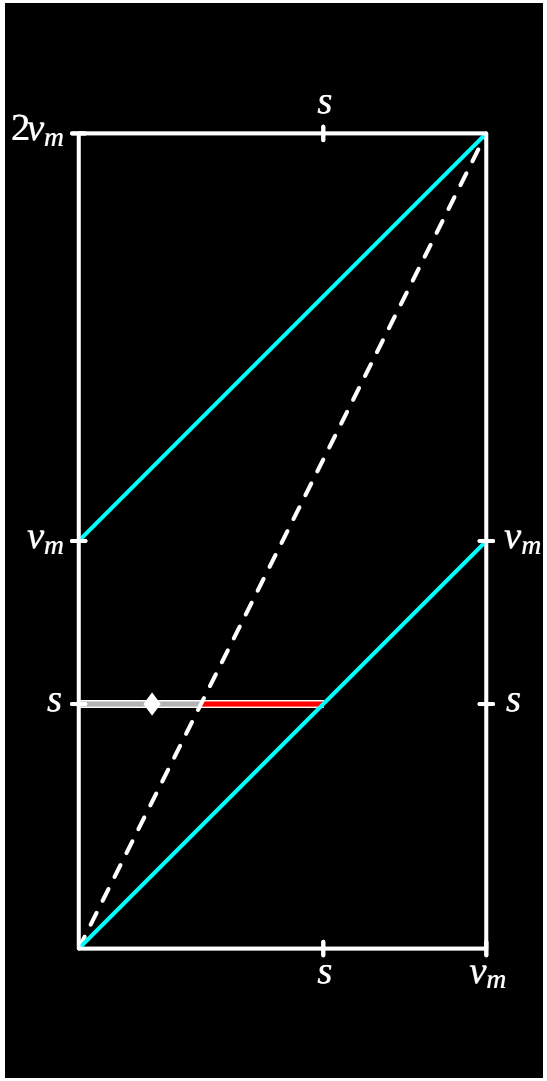
Extension map: $C_0 : s - u \mapsto u$ (maps $T_{2,2}(s)$ one-to-one onto $T_{2,1}(s)$)

Normalization: assign β_2 such as

$$\int_{T_{2,1}(s)} \beta_2(s,u)\, \mathrm{d}u = 1 \tag{0.4}$$

C_0 is **measure–conserving**: thus

$$\int_{T_{2,2}(s)} \beta_2(s, s-u)\, \mathrm{d}u = \int_{T_{2,1}(s)} \beta_2(s,u)\, \mathrm{d}u = 1. \qquad (0.5)$$


$2v_m$
s
v_m
v_m
s
s
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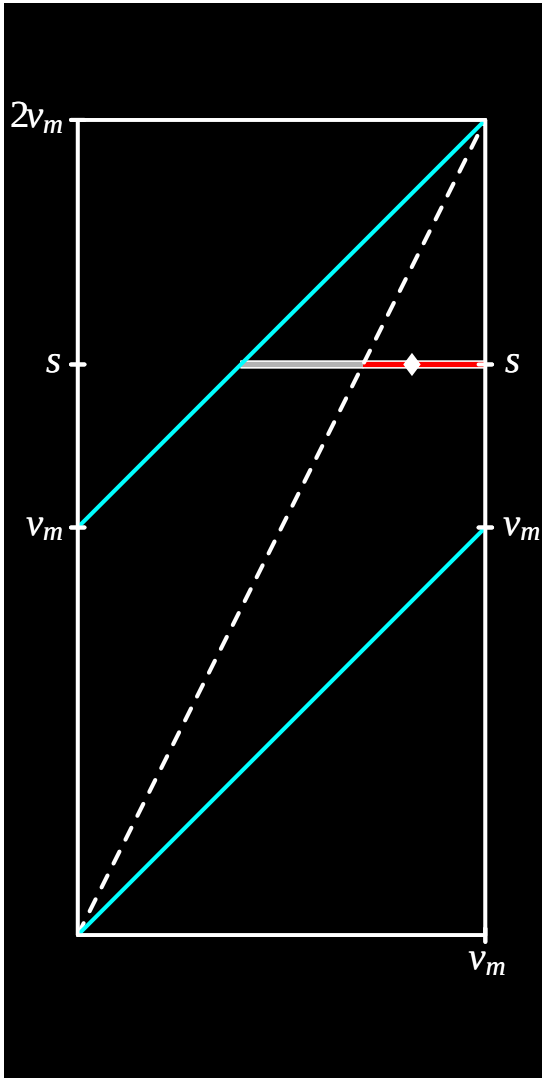
$2v_m$
s
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v_m

GENERAL CASE: $k > 2$ (multiple events)

First consider the case $s \in (0, v_m]$ and define the set of $\mathbb{R}^{k-1}$

$$T_{k,1}(s) = \left\{ 0 < u_1 \leq \ldots \leq u_{k-1} \leq s - \sum_{h=1}^{k-1} u_h \leq v_m \right\}. \qquad (0.6)$$

Clearly $\widetilde{u} = s - \sum_{h=1}^{k-1} u_h$ identifies one of the k daughters and $T_{k,1}$ is characterized by the circumstance of $\widetilde{u}$ being the volume of the *largest* daughter(s). Function β_k **is assigned** on $T_{k,1}$ in such a way that

$$\int_{T_{k,1}(s)} \beta_k (s, u_1, \ldots, u_{k-1}) \, \mathrm{d}\sigma_{k-1} = 1. \qquad (0.7)$$

We then introduce, for a fixed $s \in (0, v_m]$, the following domains in $\mathbb{R}^{k-1}$

$$T_{k,j}(s) = \left\{0 < u_1 \leq \ldots \leq u_{k-j} \leq \widetilde{u} \leq u_{k-j+1} \leq \ldots \leq u_{k-1} \leq v_m\right\},$$

$$j = 2, \ldots, k, \tag{0.8}$$

where $\widetilde{u} = s - \sum_{h=1}^{n-1} u_h$. Notice that in $T_{k,k}(s)$, $\widetilde{u}$ is the volume of the smallest drop(s).

These domains are mapped one into the other by the following measure–conserving maps:

$$C_j : (\xi_1, \ldots, \xi_{k-1}) \mapsto (u_1, \ldots, u_{k-1}), \tag{0.9}$$

$$j = 1, \ldots, k-1,$$

defined by

$$\begin{cases} u_1 = \xi_1, \ldots, u_{k-j-1} = \xi_{k-j-1}, \\ \\ u_{k-j} = s - \sum_{i=1}^{k-1} \xi_i, \\ \\ u_{k-j+1} = \xi_{k-j+1}, \ldots, u_{k-1} = \xi_{k-1}. \end{cases} \tag{0.10}$$

These maps have two main purposes:

- to “re–locate” the *residual drop* $\widetilde{u}_k$ with respect to the ordered set of the other daughters
- to extend the probability density over all domains $T_{k,j}$

Crucial (non obviuos) result: *for all* $k \geq 3$ *all open domains* $\overset{\circ}{T}_{k,j}$ *are mutually disjoint and* $\bigcap_{j=1}^{k} T_{k,j}$ *reduces to a single point which can be identified with the event*

$$u_1 = u_2 = \ldots = u_{k-1} = \frac{s}{k}, \tag{0.11}$$

that is **all droplets have the same volume**.

Because of the properties of the maps C_j, we have

$$\int_{T_{k,1}(s)} \widetilde{\beta}_k \, \mathrm{d}\sigma_{k-1} = \int_{T_{k,2}(s)} \widetilde{\beta}_k \, \mathrm{d}\sigma_{k-1} = \ldots = \int_{T_{k,k}(s)} \widetilde{\beta}_k \, \mathrm{d}\sigma_{k-1} = 1; \quad (0.12)$$

if we define $T_k = \bigcup_{j=1}^{k} T_{k,j}$ and $D_k(s,v) = T_k(s) \cap \left\{ \sum_{h=1}^{k-1} u_h = s - v \right\}$.

Thus in all the $T_{k,j}$ contributing to $D_k(s,v)$, the volume v is the one of the "residual drop". Notice that $T_k(s)$ is $(k-1)-$dimensional polytope, so that $D_k(s,v)$ **is nothing but a finite portion of an hyperplane in** $\mathbb{R}^{k-2}$.

Since we allow s in the interval $(0, 2v_m]$, **the function** β_k **is defined in a** $k-$**dimensional polytope**

$$\mathscr{T}_k = \left\{ s \in (0, 2v_m], 0 < u_1 \leq u_2 \leq \ldots \leq u_{k-1} \leq u_k, \sum_{h=1}^{k} u_h = s \right\}.$$

The domain $T_k(s)$ is just $\mathscr{T}_k \bigcap \{s = \text{constant}\}$.

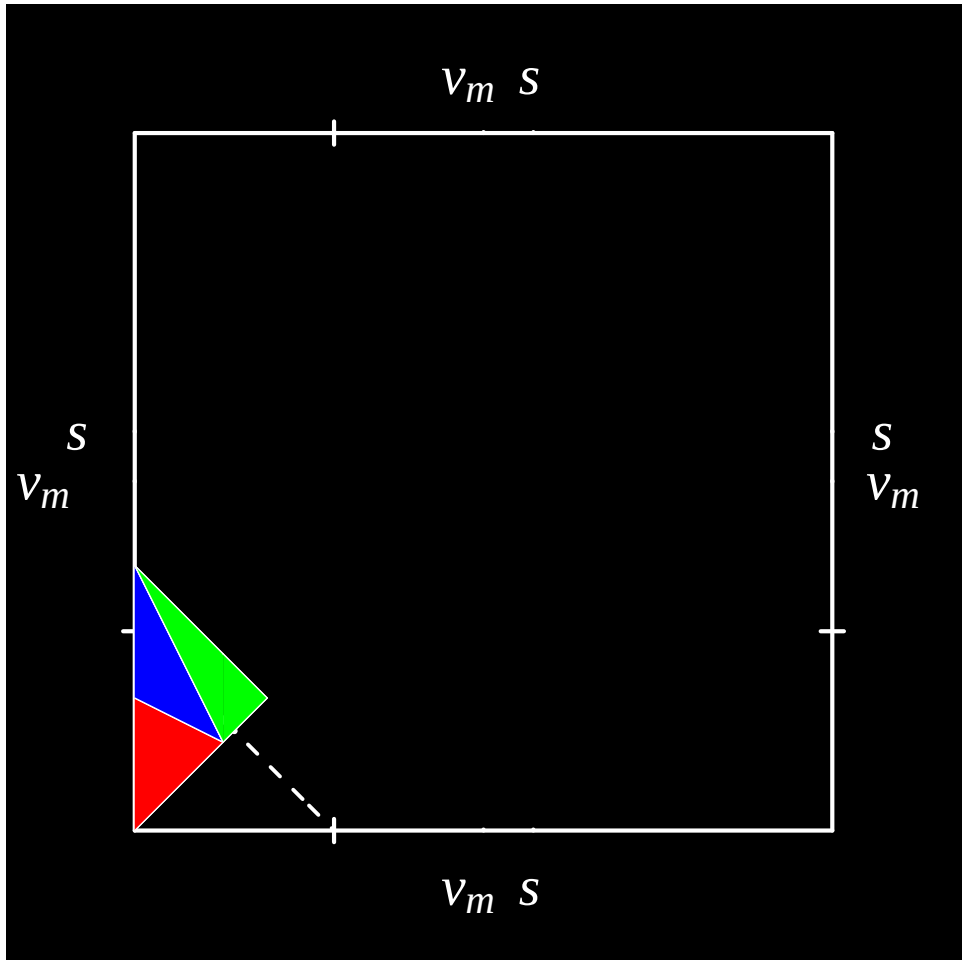


Figure 1: $s < v_m$; the stright line is $u_1 + u_2 = s - v$

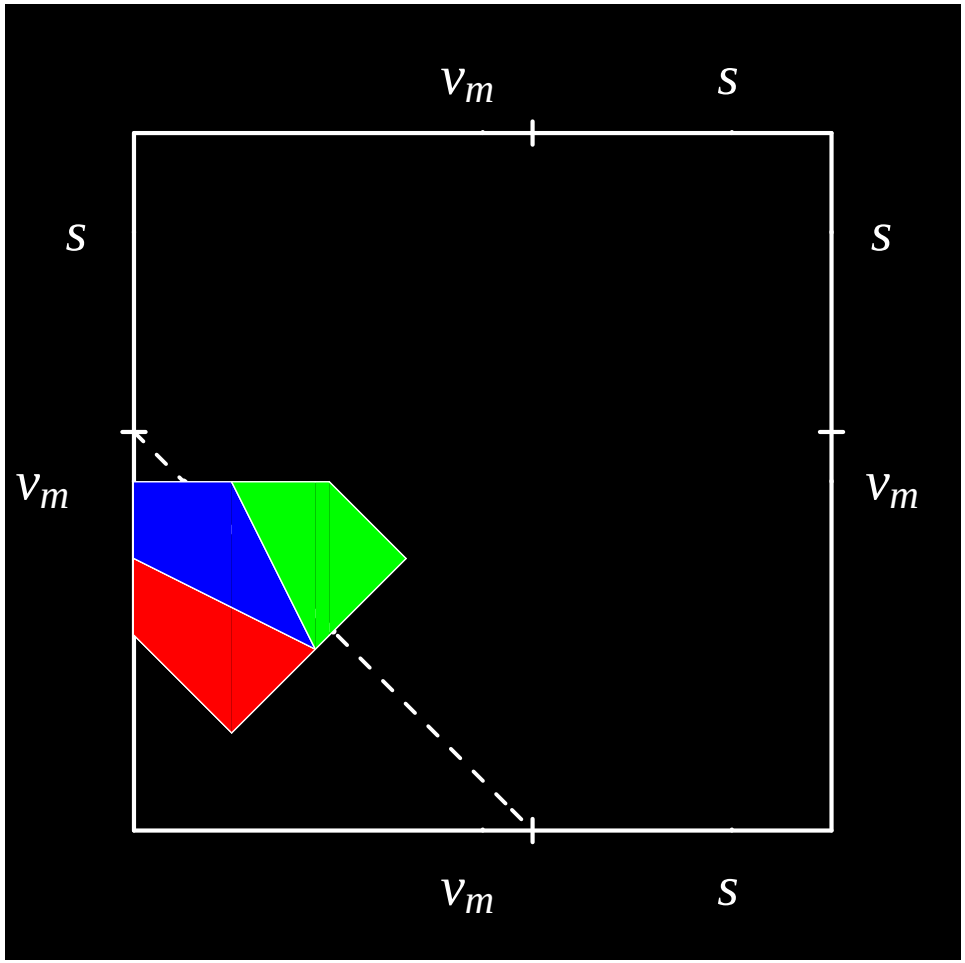


Figure 2: $s > v_m$; the stright line is $u_1 + u_2 = s - v$

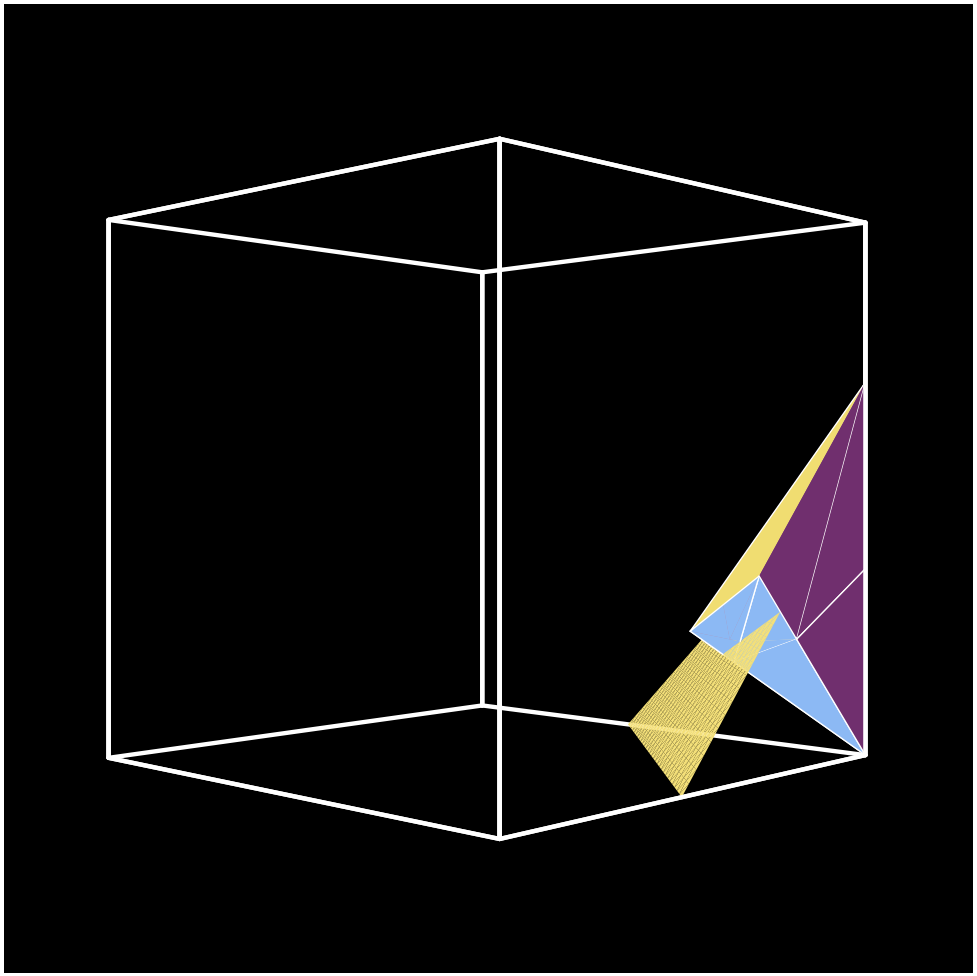


Figure 3: $s < v_m$; the intersecting plane is $u_1 + u_2 + u_3 = s - v$

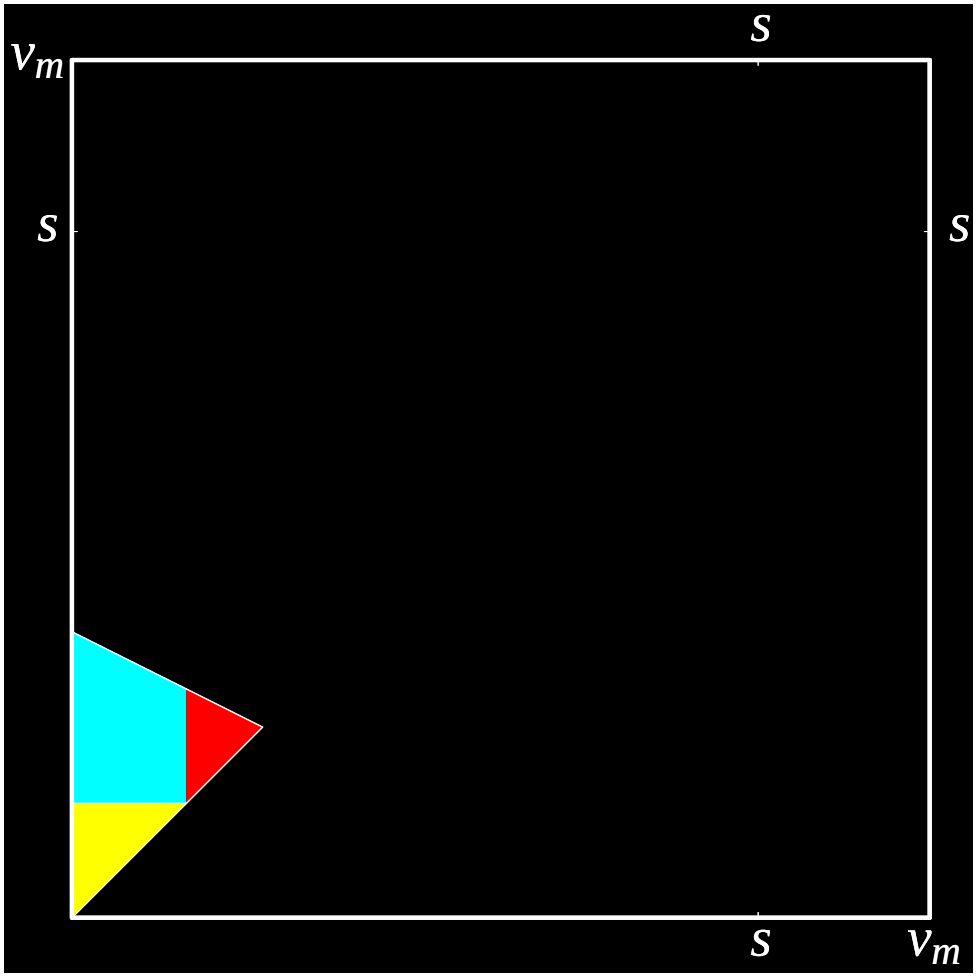


Figure 4: $s < v_m$; $u_1 u_2$ projection of the polytope–plane intersection

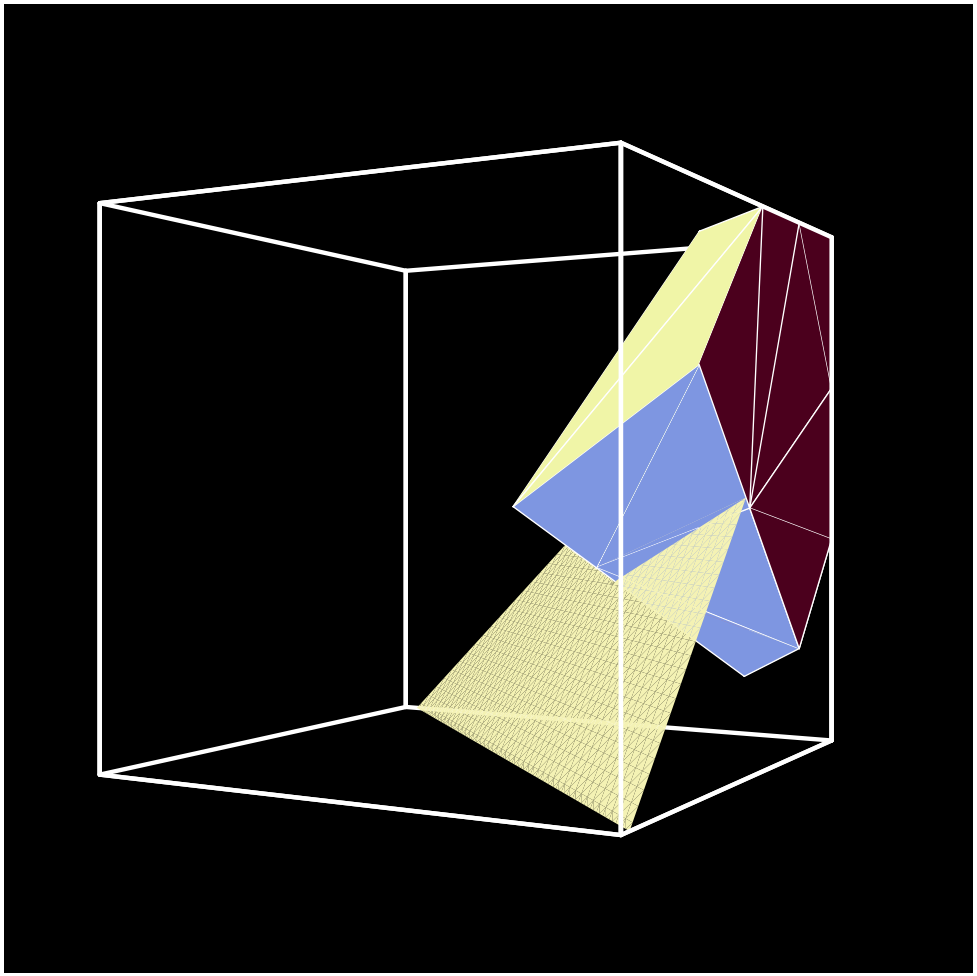


Figure 5: $s > v_m$; the intersecting plane is $u_1 + u_2 + u_3 = s - v$

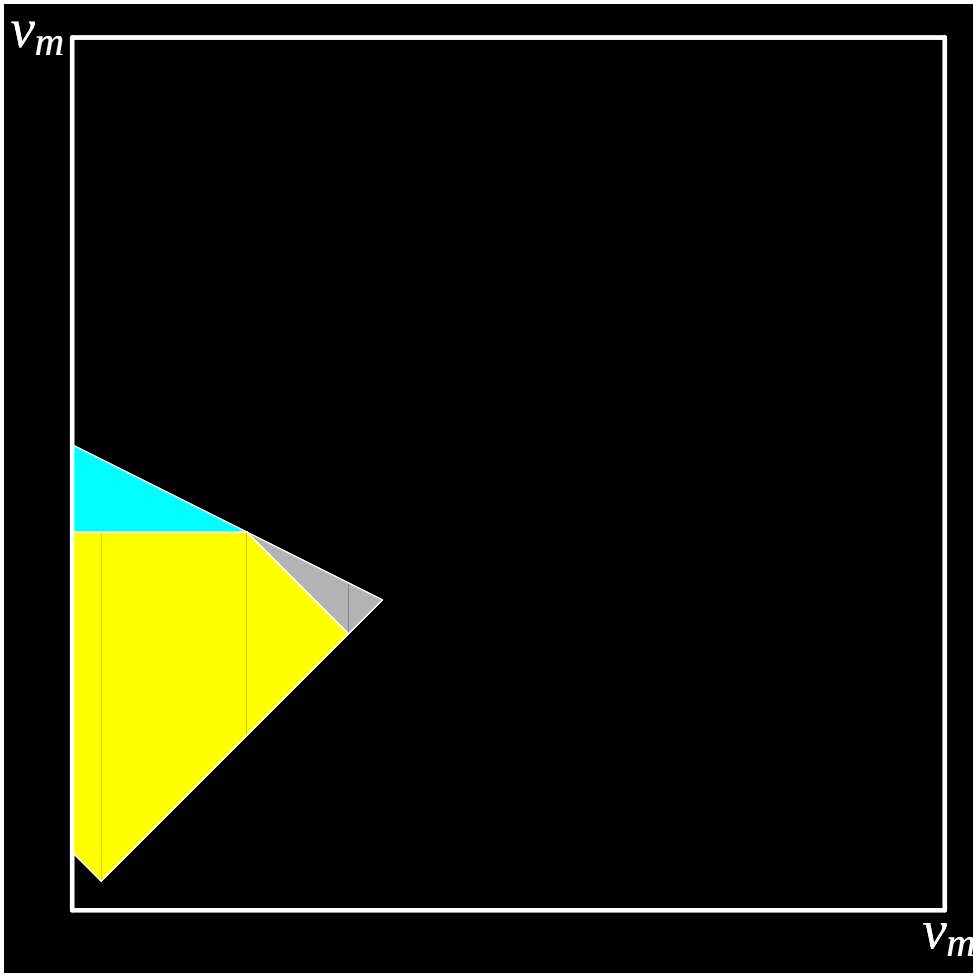


Figure 6: $s > v_m$; u_1u_2 projection of the polytope–plane intersection

EXAMPLE OF PROBABILITY FUNCTIONS

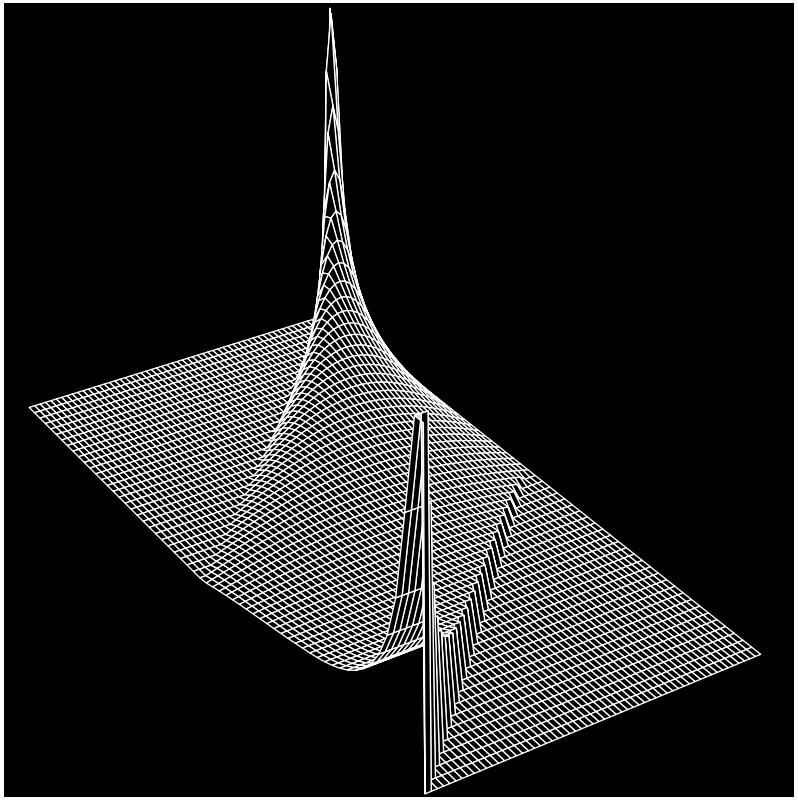


Figure 7: **Example of $\beta_2(s,u)$**

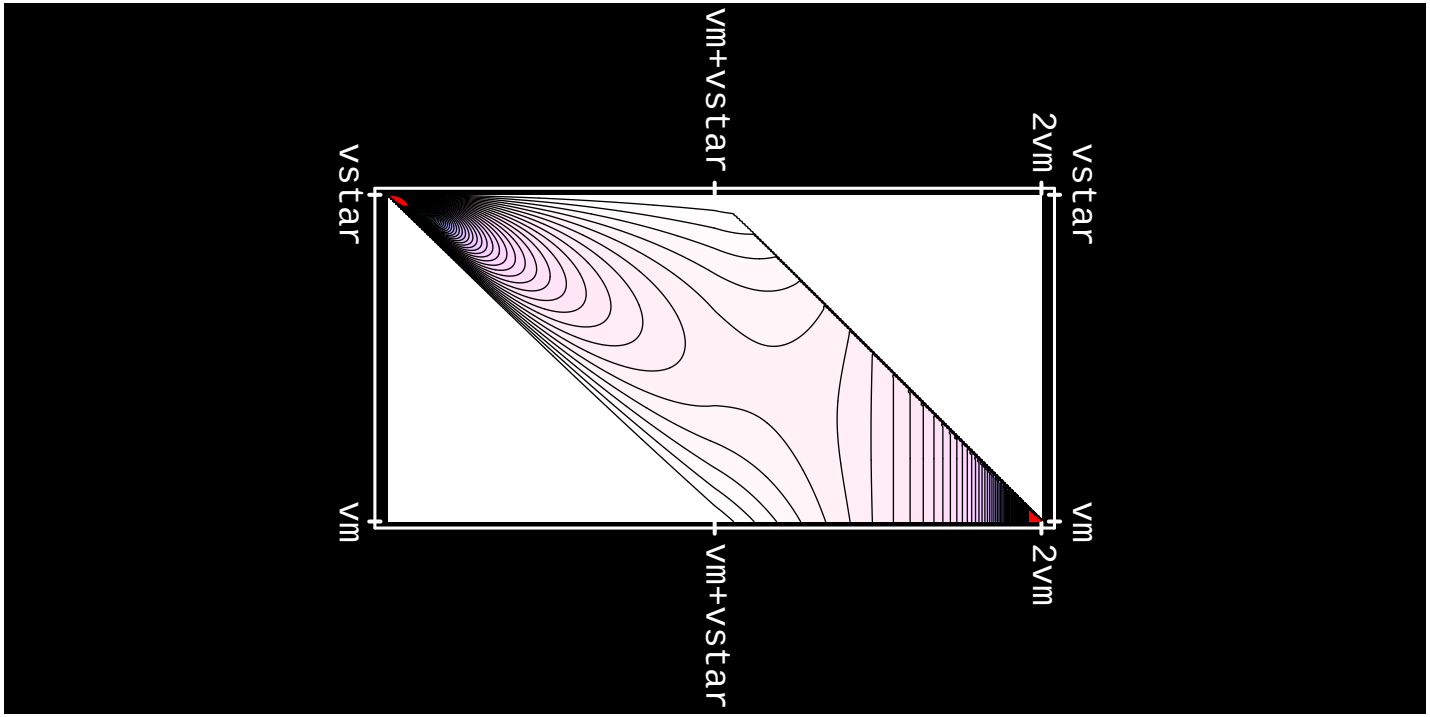
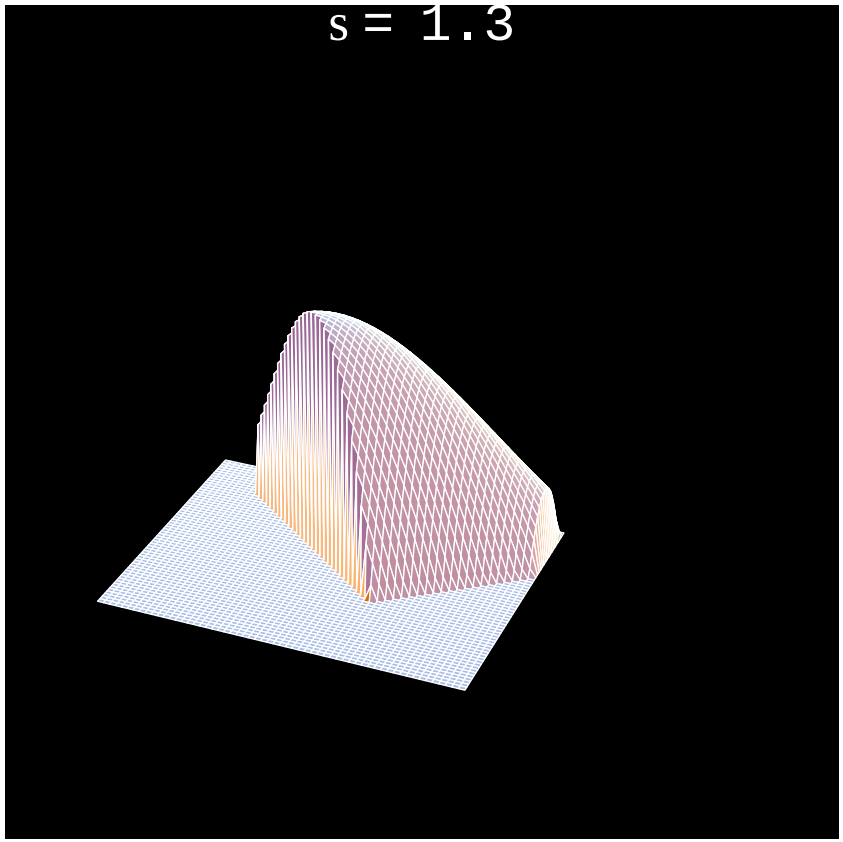


Figure 8: **Contour plot of $\beta_2(s,u)$**



Example of $\beta_3(s,u)$

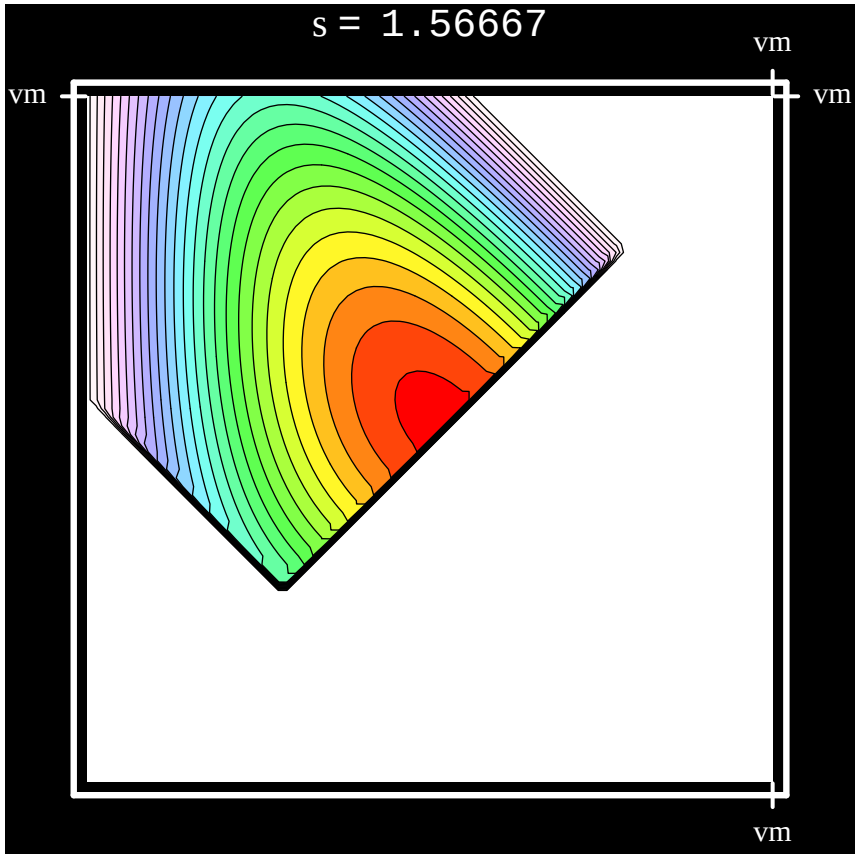


Figure 9: **Contour plot of $\beta_3(s,u)$**

Numerical Simulations

We proved existence and uniqueness globally–in–time for our model independently of how many modes are taken into account.

All simulations use the probability functions β_k shown before: events up to the forth order are taken into account. The colescence kernel is taken from the technical literature. All breakage frequency functions α_k vanish at $v = 0$ and blow up at $v = v_m$.

All multiple integral are evaluated by suitable Montecarlo Methods. For modes higher than the forth the computational time increases considerably.

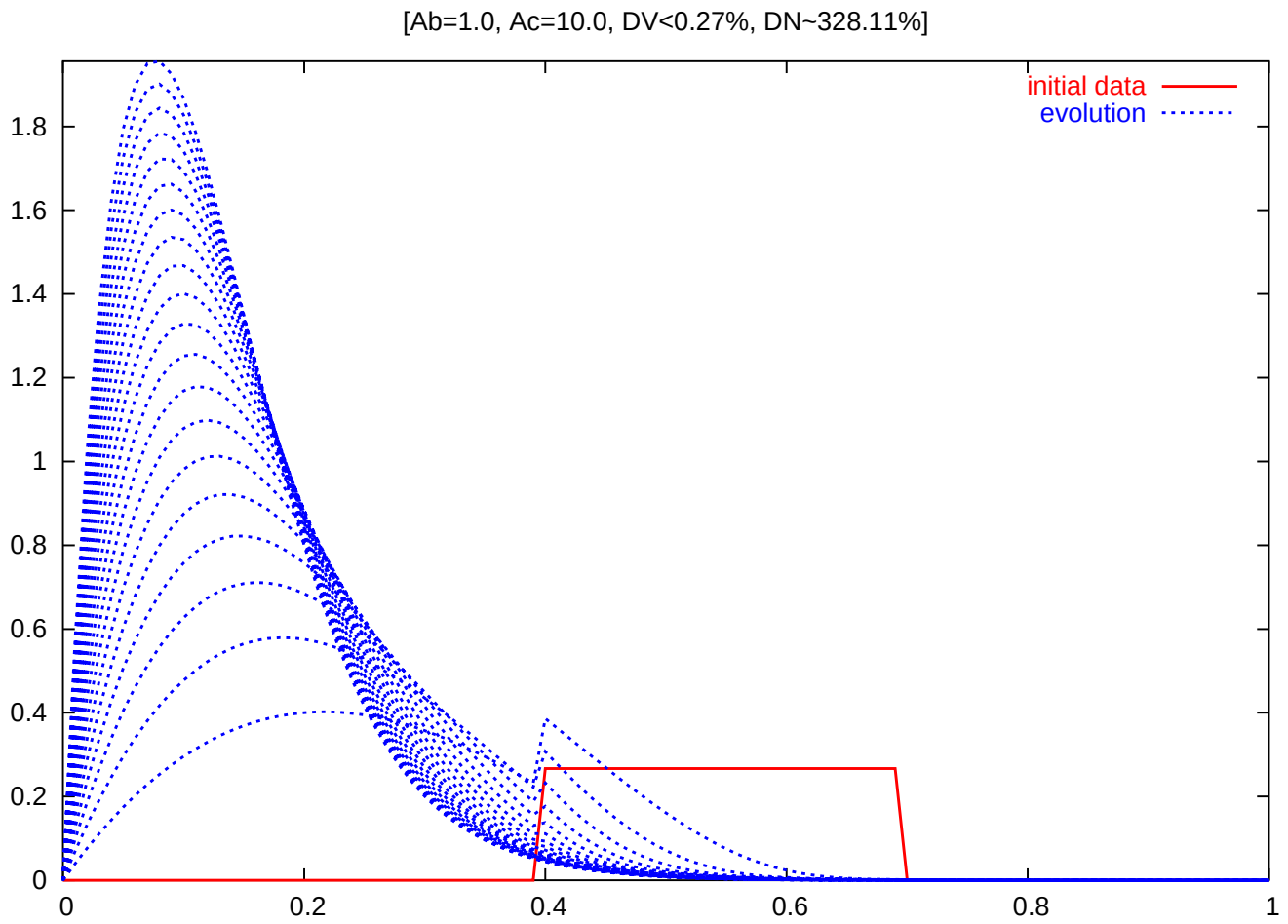


Figure 10: **Evolution of $f(v,t)$ for a given initial data: only binary events are considered**

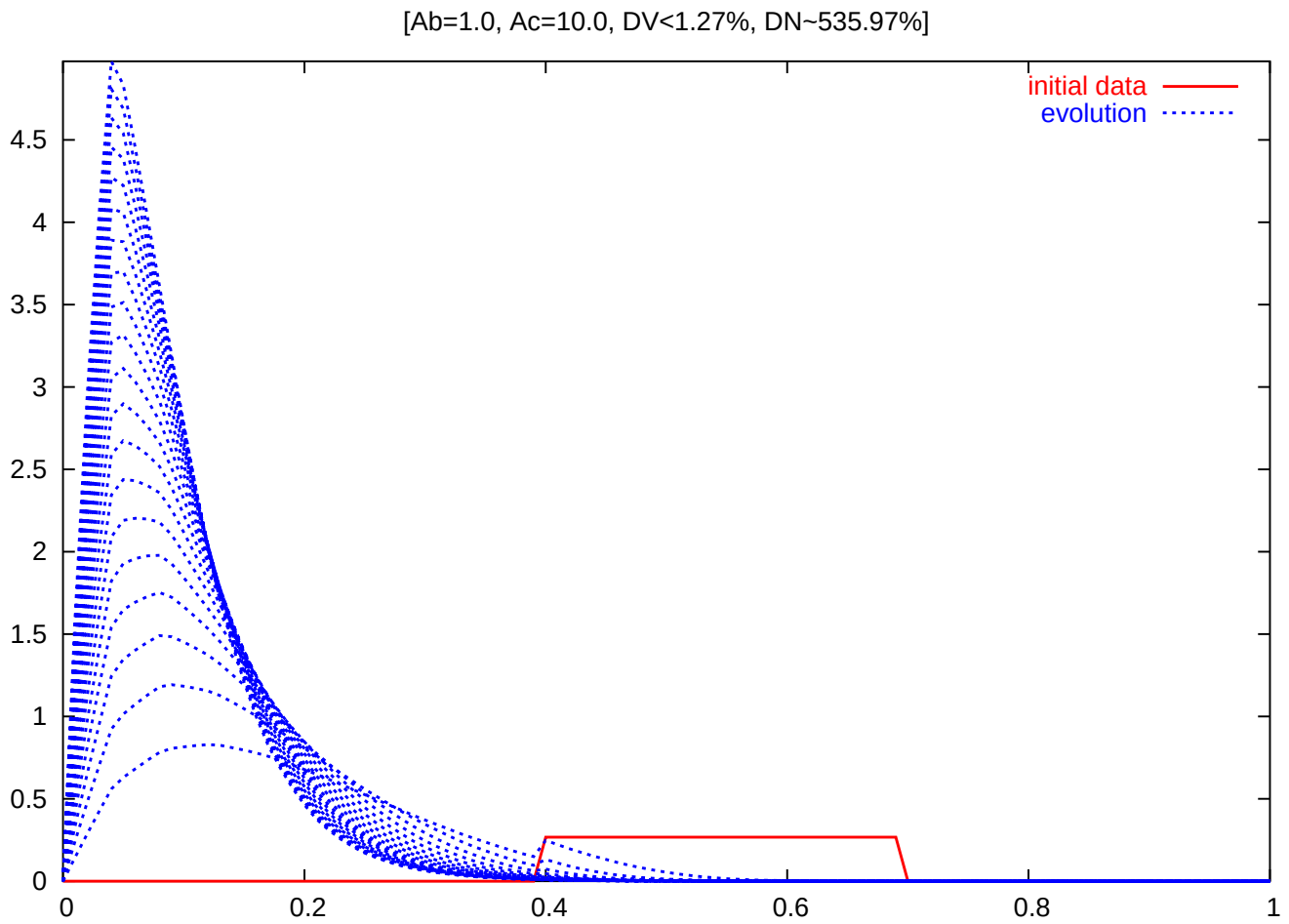


Figure 11: **Evolution of $f(v,t)$ for a given initial data : events up to the third order are considered**

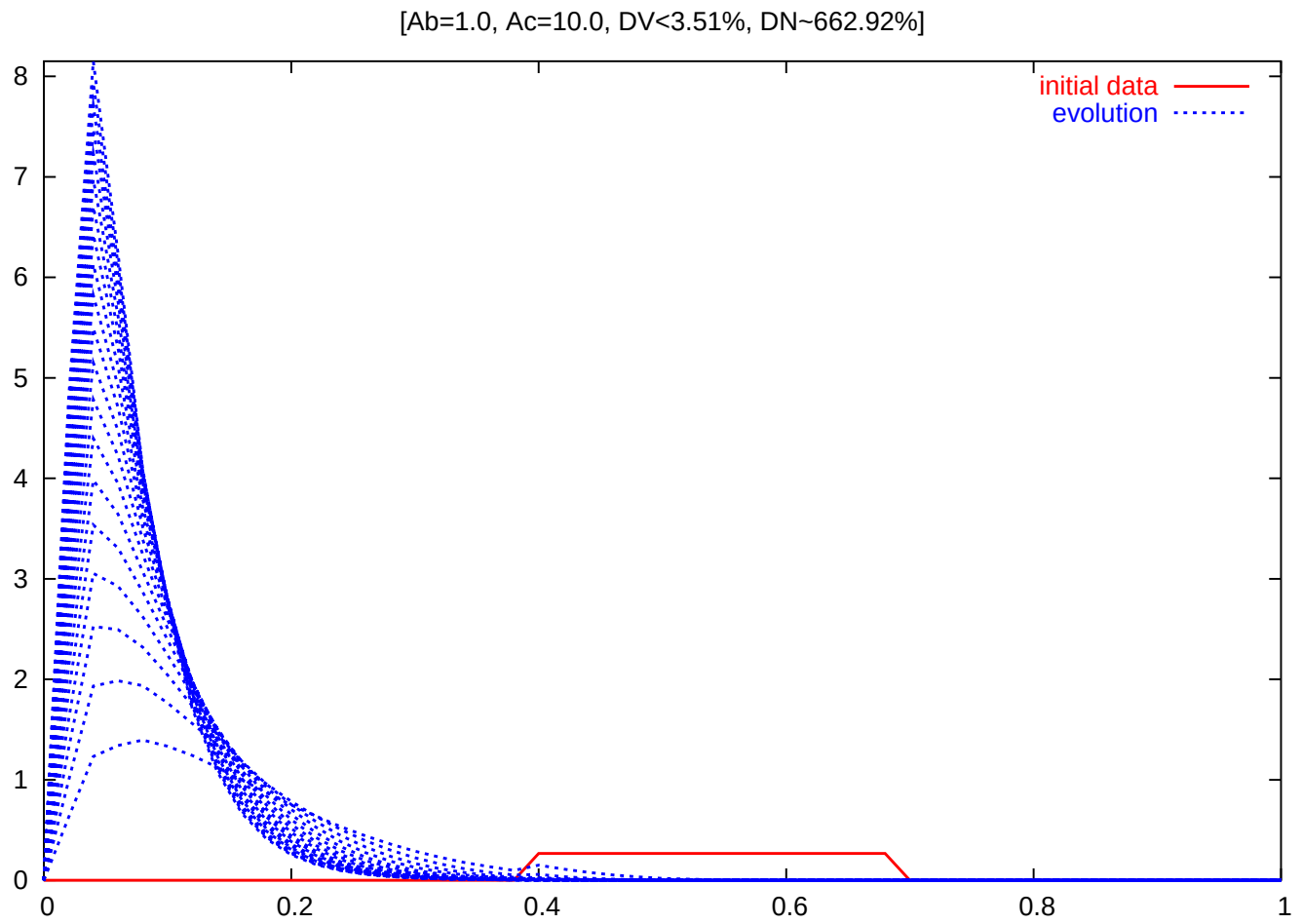


Figure 12: **Evolution of $f(v,t)$ for a given initial data : events up to the fourth order are considered**

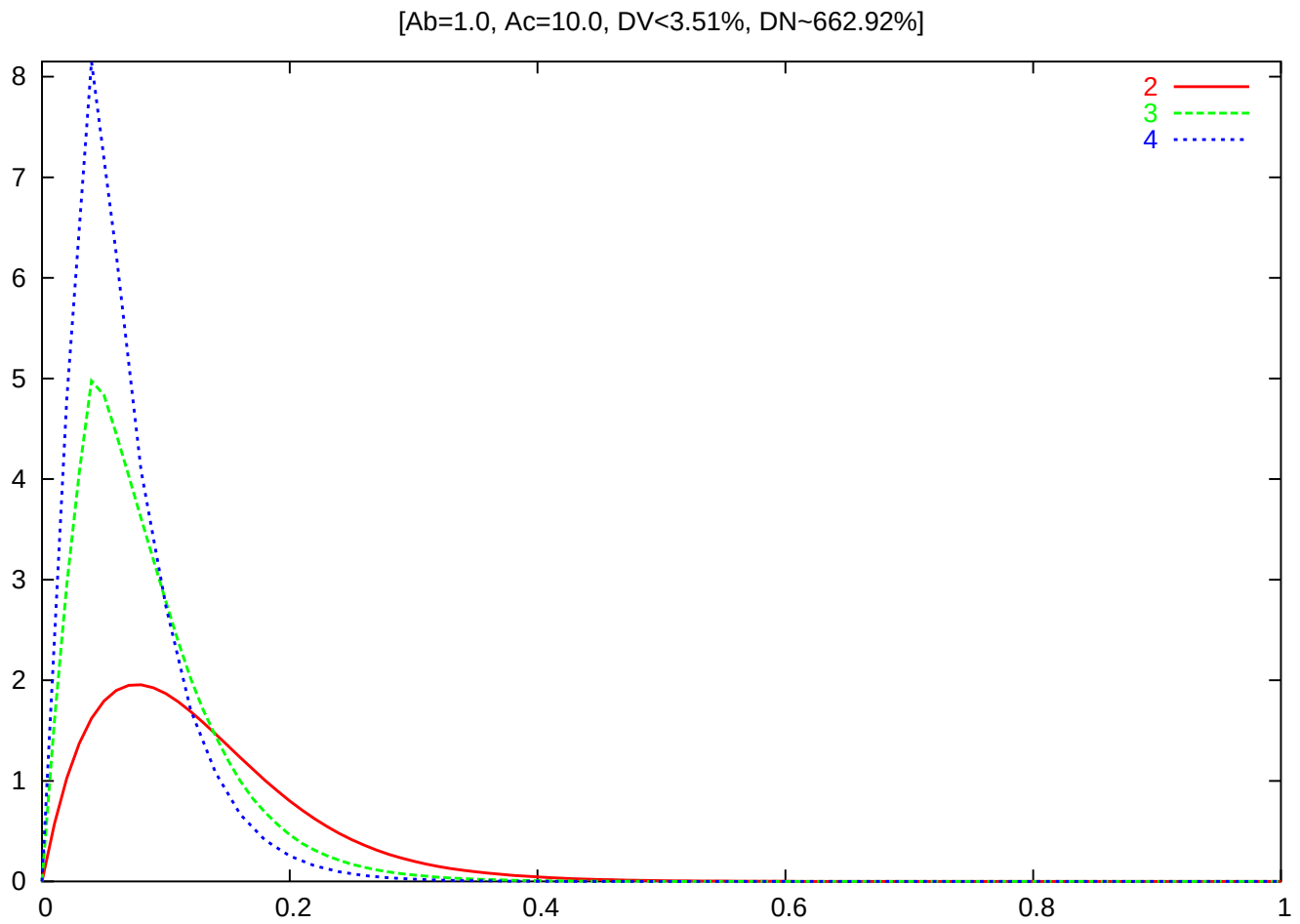


Figure 13: **Evolution of $f(v,t)$ for a given initial data: a comparison among the three previous graphs**

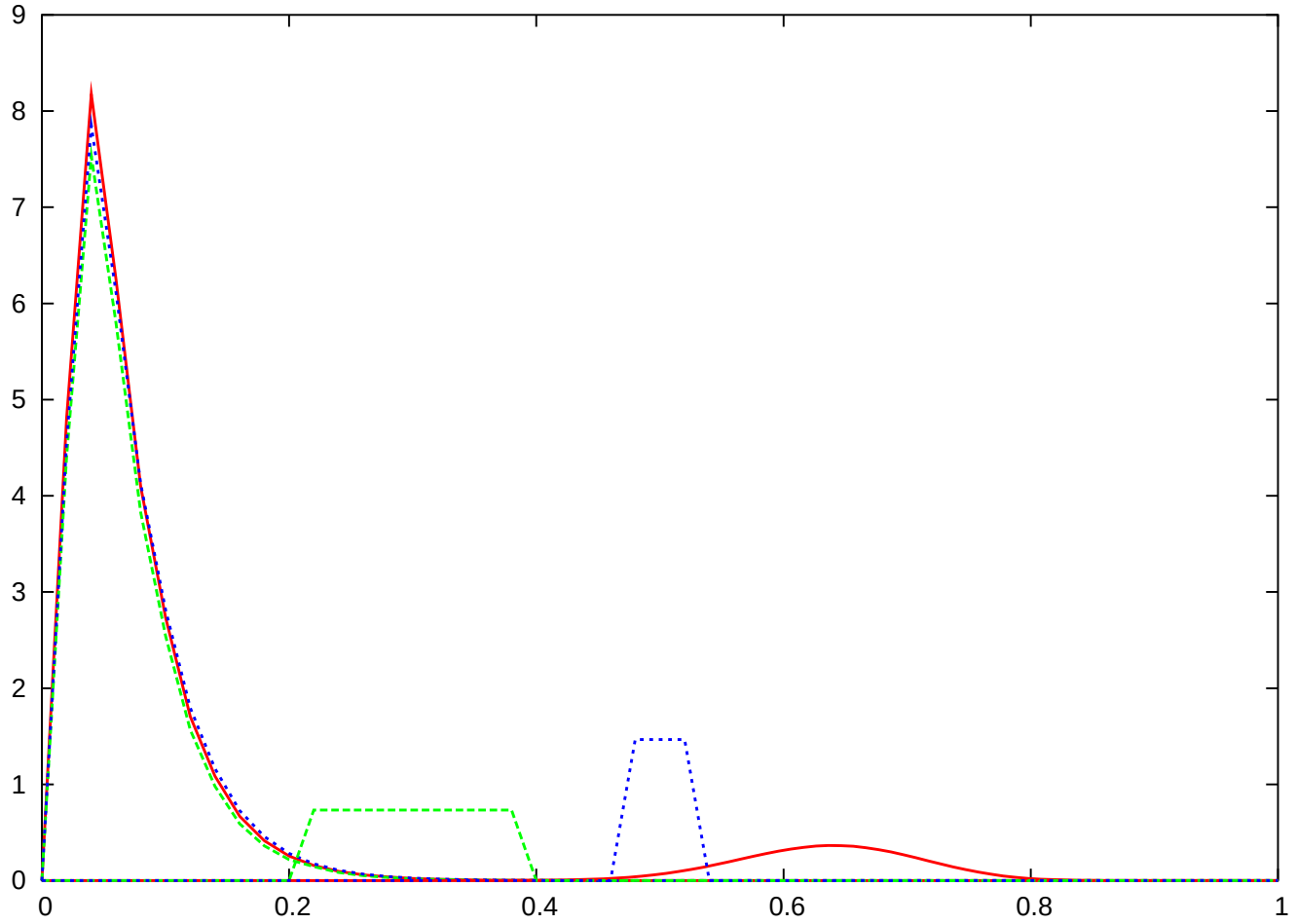


Figure 14: **Evolution of $f(v,t)$ for three different initial data: the asymptotic form is independent of $f(v,0)$**

POSSIBLE FUTURE PERSPECTIVES:

- the last graph shows that the asymptotic (in time) solution shows very little dependency upon $f_o(v)$. **Is this a theorem**?
- existence of a steady no–trivial solution has been obtained for the model with volume scattering only very recently (P. Laurençot and C. Walker 2004). **Can this be done also for the model with explicit multiple breakage** under “good” and “easy–to–read” hypotheses on $\alpha_k, \beta_k, \lambda_k$?
- Extension to the discrete case (more suitable for polymeric media) is routine for the classical model. **Is there any similar extension for our model**?
- inclusion of inlet-outlet flow (external sources), chemical agents, a temperature field....