An Interacting Diffusion Model and its Hydrodynamic Limit

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(joint work with Louis W.T. Fan)

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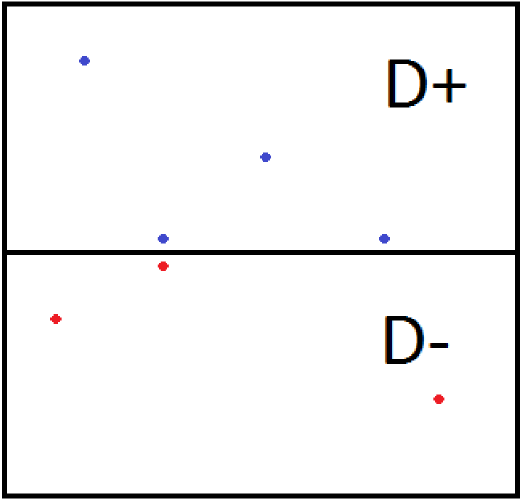
In 2008, NSF started a Solar Energy Initiative.

The purpose of the CHE-DMR-DMS Solar Energy Initiative is to support interdisciplinary efforts by groups of researchers to address the scientific challenges of highly efficient harvesting, conversion, and storage of solar energy. Groups must include three or more co-Principal Investigators; one must have demonstrated high expertise in chemistry, a second in materials research, and a third in mathematical sciences. The goal here is to create a new modality of linking the mathematical with the chemical and materials sciences to develop transformative paradigms in an area of much activity but largely incremental advances.

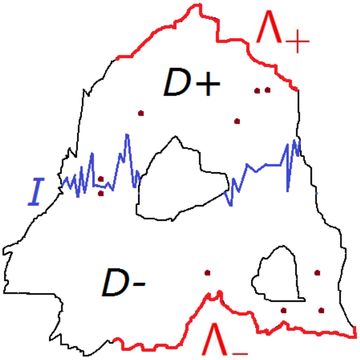
Acknowledgement: NSF grant DMR-1035196

Charge transport competes with trapping, recombination and annihilation, which greatly reduce device efficiency. It is essential to develop a comprehensive transport model that incorporate the key processes and includes nonadiabatic transitions between multiple electronic states. Most models in literature focus on statistics, and on diffusion of positive or negative charges through one type of medium. We propose a new mathematical framework, the Interacting Diffusion Model, that will describe the separation, annihilation and transport of charges in solar cells comprising multiple media and including nonadiabatic transitions between electronic states modeled (by allowing variable diffusion coefficients). A similar but simpler model of solar cells was recently reported in P. Kumar, S.C. Jain, V. Kumar, S. Chand, and R.P.A. Tandon, A model for the J-V characteristics of P3HT:PCBM solar cells. *J. Appl. Phys.* **vol. 105**, 2009.

Simplified Solar Cell



Solar Cell



Modeling

At microscopic level:

- We use reflecting Brownian motion (more generally, diffusion) with drift to model the movement of positive (and negative) charges. The drift models the electric potential these charges are subject to.
- These two types of reflecting Brownian motions with gradient drift $\frac{1}{2}\nabla(\log \rho_\pm)$ are confined within their own media and annihilate each other at certain rate when they come close near the interface, where $\rho_\pm \in C^2(\overline{D}_\pm)$ is strictly positive.
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Models

- Discrete model. Burdzy-Chen (2008, 2013): Discrete approximation of reflected Brownian motion.
- Continuous model: reflecting diffusion model.

Discrete model

- Define the grids $D_+^\varepsilon \triangleq D_+ \cap \varepsilon\mathbb{Z}^2$ and $D_-^\varepsilon \triangleq D_- \cap \varepsilon\mathbb{Z}^2$.
- Assume there are N positive charges in D_+^ε and N negative charges in D_-^ε at $t = 0$. Each particle performs continuous time simple random walk with rate $1/\varepsilon$ when $\rho_\pm = 1$ (i.e. zero potential case). For general $\rho_\pm \in C^2(\overline{D}_\pm)$, each particles performs continuous time random walk with conductance

$$\mu_{x,x+\varepsilon\vec{e}_i} \triangleq \left(1 + \frac{1}{2}\ln\frac{\rho(x+\varepsilon\vec{e}_i)}{\rho(x)}\right)\left(\frac{\rho(x)+\rho(x+\varepsilon\vec{e}_i)}{2}\right)\frac{\varepsilon^{d-2}}{2}.$$

- Any pair of particles of opposite charge is being killed with rate λ/ε if they are of distance 2ε.

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Discrete model

- Consider the configuration space

$$E^{\varepsilon} \triangleq \mathbb{N}^{D_+^{\varepsilon}} \times \mathbb{N}^{D_-^{\varepsilon}}$$

  The state of the particle system at time t will be encoded as a random element $\eta_t^{\varepsilon} = (\eta_t^{\varepsilon,+}, \eta_t^{\varepsilon,-}) \in E^{\varepsilon}$.
- $\eta^{\varepsilon,+}(x,t)$ stands for the number of positive charges at $x \in D_+^{\varepsilon}$, and $\eta^{\varepsilon,-}(x,t)$ stands for the number of negative charges at $x \in D_-^{\varepsilon}$.

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Define the empirical measures of the positive and the negative charges

$$\mathfrak{X}_t^N(dz) \triangleq \frac{1}{N} \sum_{x \in D_+^\varepsilon} \eta_t^+(x) \mathbf{1}_x(dz)$$

and

$$\mathfrak{Y}_t^N(dz) \triangleq \frac{1}{N} \sum_{x \in D_-^\varepsilon} \eta_t^-(x) \mathbf{1}_x(dz)$$

Notation: For any $S-$valued random variable X, we denote its distribution by $\mathrm{L}(X) \in M_1(S)$

Assumption

Suppose $\varepsilon \to 0$ *and* $N \to \infty$ *in such a way that* $c_1 \le N\varepsilon^2 \le c_2$ *for some constants* $0 < c_1 \le c_2 < \infty$. *Suppose* $\mathrm{L}(\mathfrak{X}_0^N) \longrightarrow \delta_{I^+(z)dz}$ *in* $M_1(M_1(\overline{D_+}))$ *with* $\|I^+\|_{L^1(D_+)} = 1$, *and* $\mathrm{L}(\mathfrak{Y}_0^N) \longrightarrow \delta_{I^-(z)dz}$ *in* $M_1(M_1(\overline{D_-}))$ *with* $\|I^-\|_{L^1(D_-)} = 1$.

Theorem

Then, as $N \to \infty$,
$\mathrm{L}((\mathfrak{X}^N, \mathfrak{Y}^N)) \longrightarrow \delta_{(\nu^+,\nu^-)} \in M_1(C([0,\infty), M_+(\overline{D_+}) \times M_+(\overline{D_-})))$
where (ν^+, ν^-) is the unique element such that
$(\nu_t^+(dz), \nu_t^-(dz)) = (u_+(t,z)\rho_+(z)dz, u_-(t,z)\rho_-(z)dz)$ *for*
$t \geq 0$, *where (u_+, u_-) satisfies the following coupled PDEs:*

$$\begin{cases} \dfrac{\partial u_+}{\partial t} = \dfrac{1}{2}\Delta u_+ + \frac{1}{2}\nabla \log \rho_+ \cdot \nabla u_+ & \text{on } (0,\infty) \times D_+, \\ \dfrac{\partial u_+}{\partial \vec{n_1}} = -\frac{\lambda}{\rho_+} u_+ u_- & \text{on } (0,\infty) \times I, \\ \dfrac{\partial u_+}{\partial \vec{n_1}} = 0 & \text{on } (0,\infty) \times (\partial D_+ \setminus I. \end{cases}$$

$$\begin{cases} \dfrac{\partial u_-}{\partial t} = \dfrac{1}{2}\Delta u_- + \frac{1}{2}\nabla \log \rho_- \cdot \nabla u_- & \text{on } (0,\infty) \times D_-, \\ \dfrac{\partial u_-}{\partial \vec{n_2}} = -\frac{\lambda}{\rho_-} u_+ u_- & \text{on } (0,\infty) \times I, \\ \dfrac{\partial u_-}{\partial \vec{n_2}} = 0 & \text{on } (0,\infty) \times (\partial D_- \setminus I. \end{cases}$$

for $t > 0$, with initial conditions $u_+(0,z) = I^+(z)$ and $u_-(0,z) = I^-(z)$. Here $\vec{n_1}$ and $\vec{n_2}$ are OUTWARD unit normals.

Outline of proof

- Establish tightness for the empirical processes:

Theorem

The sequence $\{(\mathfrak{X}^N, \mathfrak{Y}^N)\}_N$ *is relatively compact in* $\mathbb{D}((0,\infty), M_+(\overline{D_+}) \times M_+(\overline{D_-}))$. *Moreover, any limit point concentrates on* $C((0,\infty), M_+(\overline{D_+}) \times M_+(\overline{D_-}))$.

Outline of proof

- Identify the dynamics of the limiting process. Show that it is the solution of the coupled PDEs. The key is to analyze the correlation functions to show that for any fixed $t \in [0, T]$ and $\phi \in C^2(\overline{D_+})$,

$$\overline{\lim_{N\to\infty}} \mathbb{E}[\langle \mathfrak{X}_t^N, \phi\rangle] = \langle u_+(t,\cdot), \phi\rangle$$

and

$$\overline{\lim_{N\to\infty}} \mathbb{E}[(\langle \mathfrak{X}_t^N, \phi\rangle)^2] = (\langle u_+(t,\cdot), \phi\rangle)^2.$$

- Show the existence and uniqueness of solution of the coupled PDE. Probabilistic representation.

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- Show the existence and uniqueness of solution of the coupled PDE. Probabilistic representation.

Probabilistic representation

Let X be reflected BM with drift in D:

$$dX_t = dB_t + \frac{1}{2}\nabla \log \rho(X_t)dt + \vec{n}(X_t)dL_t.$$

It is a $\rho(z)dz$-symmetric diffusion on $\overline{D}$.
For $g \in \mathcal{B}_b([0,\infty) \times \partial D)$,

$$\begin{cases} \frac{\partial u}{\partial t} = \frac{1}{2}\Delta u + \frac{1}{2}\nabla(\log \rho) \cdot \nabla u & , x \in D, t > 0 \\ \frac{\partial u}{\partial \vec{n}} = \frac{1}{\rho} g u & , x \in \partial D, t > 0 \\ u(0,x) = \varphi(x) & , x \in D \end{cases}$$

has a unique solution, which can be represented by

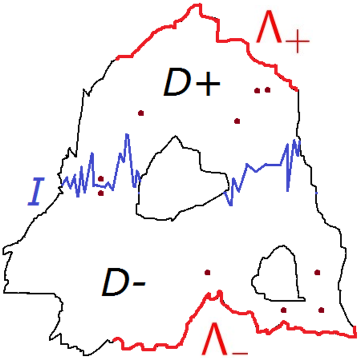
$$u(t,x) \triangleq \mathbb{E}^x[\varphi(X_t)e^{-\int_0^t g(t-s,X_s)dL_s}].$$

For the coupled PDE, for (u, v), let (Su, Sv) be the solution with u_- and u_+ in the right hand side of the equations being replaced by v and u respectively.
Using the above probabilistic representation, we can show that $S : (u, v) \mapsto (Su, Sv)$ is a contraction map when $t \leq T_0$. This yields the existence and uniqueness of the coupled PDE.

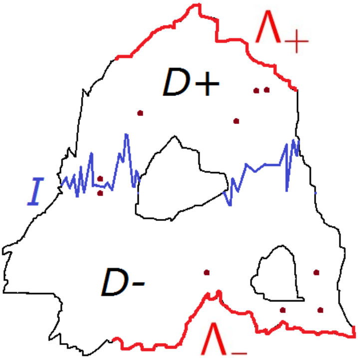
Propagation of chaos

When the number of the particles tends to infinity, they appears to be independent to each other.

Continuous model



- Particle motions in $D_\pm$ are
- $dX_t^\pm = dB_t + \frac{1}{2}\nabla(\log \rho_\pm(X_t^\pm))\,dt + \vec{n}(X^\pm)dL_t$



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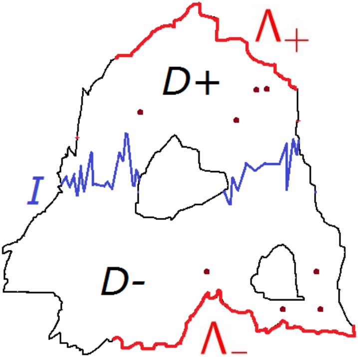

- N = number of particles, annihilation distance $\delta_N \approx N^{-1/d}$,
- annihilation rate per pair $\approx 1/\delta_N$ (given that the pair is close to interface)

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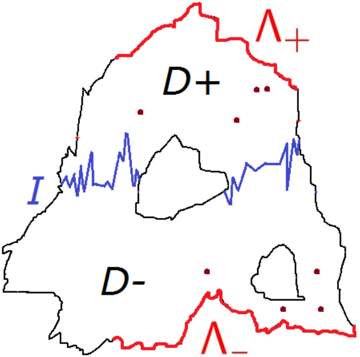

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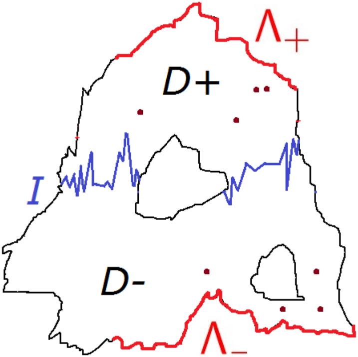

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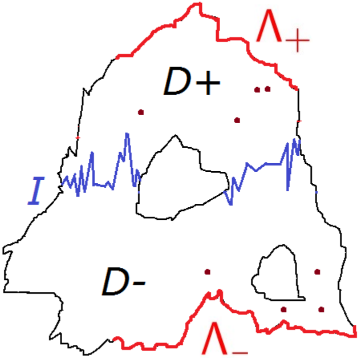

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Construction

For $\delta > 0$, define

$$I^\delta \triangleq \{(x,y) \in \overline{D}_+ \times \overline{D}_- : \sqrt{|x-z|^2 + |y-z|^2} < \delta \text{ for some } z \in I\}$$

Minkowski content:

$$\lim_{\delta \searrow 0} \frac{|I^\delta|}{c_{d+1}\,\delta^{d+1}} = \sigma(I) \quad \text{where } c_{d+1} \triangleq |\{x \in \mathbb{R}^{d+1} : |x| < 1\}|$$

Moreover,

$$\ell_\delta \triangleq \frac{1}{c_{d+1}\,\delta^{d+1}} \mathbf{1}_{I^\delta} \to \sigma\Big|_I$$

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Our model

Fix N. Define $(\mathfrak{X}_t^{+,N}, \mathfrak{X}_t^{-,N}) \in M_{\leq 1}(\overline{D}_+) \times M_{\leq 1}(\overline{D}_-)$ by

$$\Omega_N = \Omega_N^D + \Omega_N^R,$$

where

$$\Omega_N^R F(\mu) \triangleq \frac{1}{N} \sum_i \sum_j \ell_{\delta_N}(x_i, y_j) \left(F(\mu^+ - \frac{1}{n} \mathbf{1}_{\{x_i\}}, \mu^- - \frac{1}{n} \mathbf{1}_{\{y_j\}}) - F(\mu) \right)$$

if $\mu = (\frac{1}{N} \sum_i \mathbf{1}_{x_1}, \frac{1}{N} \sum_j \mathbf{1}_{y_j})$.

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Main result

Theorem

Suppose $(\mathfrak{X}_0^{+,N}, \mathfrak{X}_0^{-,N})$ *converges, and* $\liminf_{N\to\infty} N\delta_N^d > 0$.

Then

$$(\mathfrak{X}^{+,N}, \mathfrak{X}^{-,N}) \Rightarrow (u_+(t,x)\rho_+(x)dx, u_-(t,y)\rho_-(y)dy),$$

where (u_+, u_-) *is the unique solution to the following coupled heat equations:*

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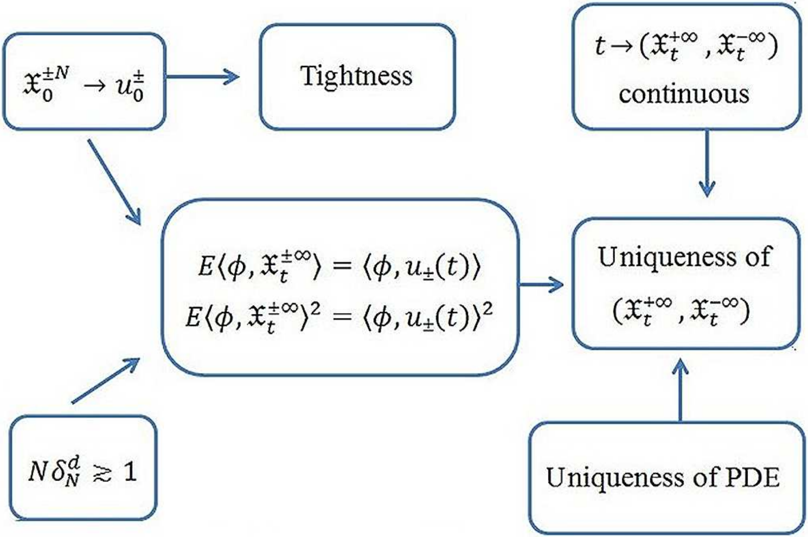
Main result: Hydrodynamic limit

Theorem

$$\begin{cases} \dfrac{\partial u_+}{\partial t} = \dfrac{1}{2}\Delta u_+ + \dfrac{1}{2}\nabla(\log \rho_+) \cdot \nabla u_+ & \textit{on } (0,\infty) \times D_+ \\ \dfrac{\partial u_+}{\partial \vec{n}_+} = \dfrac{1}{\rho_+} u_+ u_- & \textit{on } (0,\infty) \times I \\ \dfrac{\partial u_+}{\partial \vec{n}_+} = 0 & \textit{on } (0,\infty) \times (\partial D_+ \setminus I) \\ u_+ = 0 & \textit{on } (0,\infty) \times \Lambda_+ \end{cases}$$

and similar equation holds for D_-.

Idoof



$\mathfrak{X}_0^{\pm N} \to u_0^{\pm}$
Tightness
$t \to (\mathfrak{X}_t^{+\infty}, \mathfrak{X}_t^{-\infty})$ continuous
$E\langle \phi, \mathfrak{X}_t^{\pm\infty} \rangle = \langle \phi, u_{\pm}(t) \rangle$
$E\langle \phi, \mathfrak{X}_t^{\pm\infty} \rangle^2 = \langle \phi, u_{\pm}(t) \rangle^2$
Uniqueness of $(\mathfrak{X}_t^{+\infty}, \mathfrak{X}_t^{-\infty})$
$N\delta_N^d \gtrsim 1$
Uniqueness of PDE

A brief history

Rigorous derivation of macroscopic behavior of microscopic models

- Maxwell and Boltzmann's work on kinetic theory of gas
- Hilbert formulated it as a mathematical problem: his 6th problem (1900)
- Many work has been done on this subject by many people: simple exclusion process, reversible gradient system, . . . , including G.C. Papanicolaou, S.R.S. Varadhan, H.T. Yau, M.Z. Guo,
- Burdzy, Quastel (2006): N " $+$ " particles (X^i) and N " $-$ " particles performing independent RBWs. A pair of particles of opposite sign annihilates each other Simultaneously, 2 particles (one of each type) are chosen randomly to split.

What is next for our study?

- Fluctuation process $\mathfrak{Y}^{+,N}$ on D_+ defined by

$$\mathfrak{Y}_t^{+,N}(\phi) \triangleq N^{1/2}(\langle \mathfrak{X}_t^{+,N}, \phi\rangle - \mathbb{E}\langle \mathfrak{X}_t^{+,N}, \phi\rangle)$$

  where

$$\langle \mathfrak{X}_t^{+,N}, \phi\rangle \triangleq \frac{1}{N} \sum_{\alpha:\alpha\sim t} \phi(X_\alpha(t))$$

- Central limit theorem
- Large deviation

Thank you

Idoof

Lemma

As $\epsilon \to 0$, each of

$$\mathbb{E}^\infty \langle \ell_\epsilon \phi, v_+(t) \otimes v_-(t) \rangle \text{ and } \mathbb{E}\langle \ell_\epsilon \phi, \mathfrak{X}_t^{+,N} \otimes \mathfrak{X}_t^{-,N} \rangle$$

converges uniformly for $N \in \mathbb{N}$ and for any initial distribution $\{(\mathfrak{X}_0^{+,N}, \mathfrak{X}_0^{-,N})\}_N$.

Let N be the initial number of particles in each of $D_\pm$. For $n, m \in \{0, 1, 2, \dots\}$ and $t \geq 0$, we define the **correlation function** $F_t^{(n,m),N}$ by

$$\mathbb{E}\left[\frac{1}{N^{(n)}\,N^{(m)}} \sum_{\substack{i_1,\cdots i_n \\ \text{different}}}^{\sharp_t} \sum_{\substack{j_1,\cdots j_m \\ \text{different}}}^{\sharp_t} \Phi(X_t^{i_1}, \cdots, X_t^{i_n}, Y_t^{j_1}, \cdots, Y_t^{j_m})\right]$$
$$= \int_{D_+^n \times D_-^m} \Phi(\vec{x}, \vec{y})\, F_t^{(n,m),N}(\vec{x}, \vec{y})\, d(\vec{x}, \vec{y})$$

for all $\Phi \in C(\overline{D}_+^n \times \overline{D}_-^m)$, where $\sharp_t$ is the number of particles alive at time t in each of $\overline{D}_\pm$, $N^{(n)} = N(N-1)\cdots(N-n+1)$.

Theorem

$$\lim_{N\to\infty} \sup_{(t,\vec{x},\vec{y})\in[a,b]\times\overline{D}_+^n\times\overline{D}_-^m} F_t^{(n,m),N}(\vec{x},\vec{y}) = \prod_{i=1}^{n} u_+(t,x_i) \prod_{j=1}^{m} u_-(t,y_j)$$

Idoof:

- $\{F^{(n,m),N}\}_N$ is equi-continuous and uniformly bounded
- Let $F^{(n,m),\infty}$ be a subsequential limit, then
- both $F^{(n,m),\infty}$ and $\prod_{i=1}^n u_+(t,x_i) \prod_{j=1}^m u_-(t,y_j)$ satisfy the same infinite system of hierarchical equations
- Uniqueness of solution for the hierarchy

$$
\begin{aligned}
& F_t^{(n,m)}(\vec{r},\vec{s}) \\
= & \int_{D_+^n \times D_-^m} \Phi^{(n,m)}(\vec{a},\vec{b})\, p_t((\vec{r},\vec{s}),(\vec{a},\vec{b}))\, d(\vec{a},\vec{b}) \\
& - \int_{\theta=0}^{t} \Bigg(\sum_{i=1}^{n} \int_{\partial_+^i} F_\theta^{(n,m+1)}(\vec{a},(\vec{b},a_i))\, p_{t-\theta}((\vec{r},\vec{s}),(\vec{a},\vec{b}))\, d\sigma(\vec{a},\vec{b}) \\
& \qquad \sum_{j=1}^{m} \int_{\partial_-^j} F_\theta^{(n+1,m)}((\vec{a},b_j),\vec{b}))\, p_{t-\theta}((\vec{r},\vec{s}),(\vec{a},\vec{b}))\, d\sigma(\vec{a},\vec{b}) \Bigg)
\end{aligned}
$$

where σ is the surface measure of $\partial(D_+^n \times D_-^m)$, $\Phi^{(n,m)}(\vec{a},\vec{b}) \triangleq \prod_{i=1}^n u_0^+(a_i) \prod_{j=1}^m u_0^-(b_j)$ and

$$\partial_+^i \triangleq \big(D_+ \times \cdots \times \overset{i^{th}}{(\partial D_+ \cap I)} \times \cdots \times D_+\big) \times D_-^m \qquad (0.1)$$

$$\partial_-^j \triangleq D_+^n \times \big(D_- \times \cdots \times \overset{j^{th}}{(\partial D_- \cap I)} \times \cdots \times D_-\big) \qquad (0.2)$$