Exponential Convergence to Equilibrium for Nonlinear Reaction-Diffusion Systems Arising in Reversible Chemistry

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Abstract. We consider a prototypical nonlinear reaction-diffusion system arising in reversible chemistry. Based on recent existence results of global weak and classical solutions derived from entropy-decay related a-priori estimates and duality methods, we prove exponential convergence of these solutions towards equilibrium with explicit rates in all space dimensions.

The key step of the proof establishes an entropy entropy-dissipation estimate, which relies only on natural a-priori estimates provided by mass-conservation laws and the decay of an entropy functional.

Keywords: Reaction-diffusion equations · Entropy method · Duality method · Large-time behaviour · Convergence to equilibrium

1 Introduction

Reaction-Diffusion Systems for Reversible Chemistry

The evolution of a mixture of diffusive species $\mathcal{A}_i, i = 1, 2, \ldots, q$, undergoing a reversible reaction of the type

$$\alpha_1 \mathcal{A}_1 + \cdots + \alpha_q \mathcal{A}_q \rightleftharpoons \beta_1 \mathcal{A}_1 + \cdots + \beta_q \mathcal{A}_q, \qquad \alpha_i, \beta_i \in \mathbb{N},$$

is modelled using mass-action kinetics (see e.g. [3–5,9] for a derivation from basic principles) in the following way:

$$\partial_t a_i - d_i\, \Delta_x a_i = (\beta_i - \alpha_i)\left(l \prod_{j=1}^{q} a_j^{\alpha_j} - k \prod_{j=1}^{q} a_j^{\beta_j} \right), \tag{1}$$

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where $a_i := a_i(t,x) \geq 0$ denotes the concentration at time t and point x of the species A_i and $d_i > 0$ are positive and constant diffusion coefficients.

We suppose that $x \in \Omega$, where Ω is a bounded domain of $\mathbb{R}^N$ $(N \geq 1)$ with sufficiently smooth (e.g. $C^{2+\alpha}$, $\alpha > 0$) boundary $\partial\Omega$, and complement system (1) by homogeneous Neumann boundary conditions:

$$n(x) \cdot \nabla_x a_i(t,x) = 0\,, \qquad \forall t \geq 0,\ x \in \partial\Omega, \tag{2}$$

where $n(x)$ is the outer normal unit vector at point x of $\partial\Omega$.

The particular case $\mathcal{A}_1 + \mathcal{A}_2 \rightleftharpoons \mathcal{A}_3 + \mathcal{A}_4$ (that is, when $q = 4$ with $\alpha_1 = \alpha_2 = 1$, $\beta_3 = \beta_4 = 1$, $\alpha_3 = \alpha_4 = 0$ and $\beta_1 = \beta_2 = 0$) has lately received a lot of attention as a prototypical model system featuring quadratic nonlinearities, see e.g. [7,12,17]. For the sake of readability, we shall set $l = 1 = k$ (the general case can be treated without any additional difficulty) and assume that Ω is normalised (i.e. $|\Omega| = 1$). We then consider the particular case of system (1), which writes as

$$\begin{cases} \partial_t a_1 - d_1\, \Delta_x a_1 = a_3\, a_4 - a_1\, a_2, \\ \partial_t a_2 - d_2\, \Delta_x a_2 = a_3\, a_4 - a_1\, a_2, \\ \partial_t a_3 - d_3\, \Delta_x a_3 = a_1\, a_2 - a_3\, a_4, \\ \partial_t a_4 - d_4\, \Delta_x a_4 = a_1\, a_2 - a_3\, a_4, \end{cases} \tag{3}$$

together with the homogeneous Neumann boundary conditions (2).

It was first proven by Goudon and Vasseur in [17] based on an intricate use of De Giorgi's method that whenever d_1, d_2, d_3, $d_4 > 0$, there exists a global smooth solution for dimensions $N = 1, 2$. For higher space dimensions the existence of classical solutions constitutes an open problem, for which the Hausdorff dimension of possible singularities was characterised in [17]. The (technical) criticality of quadratic nonlinearities was underlined by Caputo and Vasseur in [8], where smooth solutions were shown to exist in any dimension for systems with a nonlinearity of power law type which is strictly subquadratic, see also e.g. [1].

A further related result by Hollis and Morgan [20] showed that if blow-up (here that is a concentration phenomena since the total mass is conserved) occurs in one concentration $a_i(t,x)$ at some time t and position x, then at least one more concentration has to blow-up (i.e. concentrate) at the same time and position. A proof of these results is based on a duality argument.

In [12], a duality argument in terms of entropy density variables was used to prove in an elegant way the existence of global L^2-weak solutions in any space dimension. Recently in [7], a nice improvement of the duality methods allows to show global classical solutions in 2D of the prototypical system (3)–(2) in a significantly shorter and less technical way than via De Giorgi's method.

In the present work, we shall show that exponential convergence (with explicit rates) towards the unique constant equilibrium still holds for any dimension N (see Theorem 1 below) when one considers L^2-weak solutions. The proof of Theorem 1 is based on an approach, where a quantitative entropy entropy-dissipation estimate is established, which uses only natural a-priori bounds of the system, and thus significantly improves the results of [11] and related previous results like [10,15,16,18].

The paper is organized as follows: We start in Sect. 2 by presenting a-priori bounds for our system and by overviewing the available analytical tools. Next, in Sect. 3, we prove Theorem 1 stating exponential convergence to equilibrium.

2 A Priori Estimates and Analytical Tools

2.1 Mass Conservation Laws

The conservation of the number of atoms implies (at first for all smooth solutions $(a_i)_{i=1,..,4}$ of (3) with Neumann condition (2)) that for all $t \geq 0$,

$$\begin{cases} M_{13} := \int_\Omega (a_1(t,x) + a_3(t,x))\, dx = \int_\Omega (a_1(0,x) + a_3(0,x))\, dx, \\ M_{14} := \int_\Omega (a_1(t,x) + a_4(t,x))\, dx = \int_\Omega (a_1(0,x) + a_4(0,x))\, dx, \\ M_{23} := \int_\Omega (a_2(t,x) + a_3(t,x))\, dx = \int_\Omega (a_2(0,x) + a_3(0,x))\, dx, \\ M_{24} := \int_\Omega (a_2(t,x) + a_4(t,x))\, dx = \int_\Omega (a_2(0,x) + a_4(0,x))\, dx. \end{cases} \tag{4}$$

Note that only three of the above four conservation laws are linearly independent.

2.2 Entropy Functional and Entropy Dissipation

A second set of a-priori estimates stems from the nonnegative entropy (free energy) functional $E((a_i)_{i=1,..,4})$ and the entropy dissipation $D((a_i)_{i=1,..,4}) = -\frac{d}{dt}E((a_i)_{i=1,..,4})$ associated to (3):

$$E(a_i(t,x)_{i=1,..,4}) = \sum_{i=1}^{4} \int_\Omega \Big(a_i(t,x)\log(a_i(t,x)) - a_i(t,x) + 1 \Big)\, dx, \tag{5}$$

$$\begin{aligned} D(a_i(t,x)_{i=1,..,4}) = &\sum_{i=1}^{4} \int_\Omega 4\, d_i\, |\nabla_x \sqrt{a_i(t,x)}|^2\, dx \\ &+ \int_\Omega (a_1\, a_2 - a_3\, a_4) \log\left(\frac{a_1\, a_2}{a_3\, a_4}\right)(t,x)\, dx. \end{aligned} \tag{6}$$

It is easy to verify that the following entropy dissipation law holds (still for sufficiently regular solutions $(a_i)_{i=1,..,4}$ of (3) with (2)) for all $t \geq 0$

$$E(a_i(t,x)_{i=1,..,4}) + \int_0^t D(a_i(s,x)_{i=1,..,4})\, ds = E(a_i(0,x)_{i=1,..,4})\,. \tag{7}$$

The entropy decay estimate (7) implies as a first a-priori estimate that

$$a_i \in L^\infty([0,+\infty[; L\log L(\Omega)), \qquad \forall i = 1,..,4\,. \tag{8}$$

Considering in (7) that the time integral of the entropy dissipation (6) is uniformly bounded-in-time, its first component provides the estimate

$$\sqrt{a_i} \in L^2([0,+\infty[; H^1(\Omega)), \qquad \forall i = 1,..,4, \tag{9}$$

Finally, the second component of the time integral of the entropy dissipation (6) ensures that, provided that $a_3\, a_4 \in L^1_{loc}([0,+\infty[\times\bar{\Omega})$, then also $a_1\, a_2 \in L^1_{loc}([0,+\infty[\times\bar{\Omega})$. This comes out of the following classical inequality (cf. [14]), which holds for any $\kappa > 1$,

$$a_1\, a_2 \le \kappa\, a_3\, a_4 + \frac{1}{\log \kappa}\,(a_1\, a_2 - a_3\, a_4)\, \log\left(\frac{a_1\, a_2}{a_3\, a_4}\right). \tag{10}$$

Note that by letting κ be as large as necessary, this inequality also allows to prove that an approximating sequence $a_1^n\, a_2^n$ is (locally in time) weakly compact in L^1 if the sequence $a_3^n\, a_4^n$ is also weakly compact in L^1 (and when estimate (7) holds uniformly with respect to n).

Remark 1. *We remark (see [12]), that as a consequence of the first two entropy related a-priori estimates* (8)–(9)*, global classical solutions of system* (3)–(2) *can be constructed only in 1D. In 2D, global L^2-weak solutions can be deduced by using Trudinger's inequality. In any higher space dimension, renormalised solution can be obtained from all three a-priori estimate* (8)–(10)*.*

2.3 Entropy Structure and Duality Methods

The system (3)–(2) can also be rewritten in terms of the entropy density variables $z_i := a_i \log(a_i) - a_i$. By introducing the sum $z := \sum_{i=1}^4 z_i$, it holds that

$$\begin{cases} \partial_t z - \Delta_x\,(A\, z) \le 0, \qquad n(x)\cdot\nabla_x z_i(t,x) = 0, \\ A(t,x) := \frac{\sum_{i=1}^4 d_i\, z_i}{\sum_{i=1}^4 z_i} \in \Big[\min\limits_{i=1,..,4}\{d_i\},\, \max\limits_{i=1,..,4}\{d_i\}\Big], \end{cases} \tag{11}$$

Then, by a duality argument (see e.g. [12, 20, 21] and the references therein), the parabolic problem (11) satisfies for all $T > 0$ and $\Omega_T = (0,T)\times\Omega$ and for all space dimensions $N \ge 1$ the following a-priori estimate

$$\|z_i\|_{L^2(\Omega_T)} \le C(1+T)^{1/2} \left\| \sum_{i=1}^4 a_{i0}(\log(a_{i0}) - 1)\right\|_{L^2(\Omega)}, \qquad i = 1,..,4, \tag{12}$$

where C is a constant independent of T, see [7, 12]. Thus, given $(a_{i0})_{i=1,..,4} \in L^2(\log L)^2(\Omega)$, we have $(a_i)_{i=1,..,4} \in L^2(\log L)^2(\Omega_T)$ and the quadratic nonlinearities on the right hand side of (3) are uniformly integrable, which allows to prove the existence of global L^2-weak solutions in all space dimensions $N \ge 1$ [12]. Moreover, in 2D and in higher space dimension under the assumption of sufficiently "similar" diffusion coefficients (i.e. $\max\{d_i\} - \min\{d_i\}$ is sufficiently small), an improved duality estimate allows to show global classical solutions [7].

2.4 Equilibrium

We observe that when all the diffusivity constants $(d_i)_{i=1,..,4} > 0$ are positive, there exists a unique constant equilibrium state $(a_{i,\infty})_{i=1,..,4}$ (for which the entropy dissipation vanishes). It is defined by the unique positive constants balancing the reversible reaction $a_{1,\infty}\, a_{2,\infty} = a_{3,\infty}\, a_{4,\infty}$ and satisfying the conservation laws $a_{j,\infty} + a_{k,\infty} = M_{jk}$ for $(j,k) \in (\{1,2\},\{3,4\})$, that is:

$$\begin{cases} a_{1,\infty} = \frac{M_{13}M_{14}}{M}\,, \quad & a_{3,\infty} = M_{13} - \frac{M_{13}M_{14}}{M} = \frac{M_{13}M_{23}}{M}\,, \\ a_{2,\infty} = \frac{M_{23}M_{24}}{M}\,, \quad & a_{4,\infty} = M_{14} - \frac{M_{13}M_{14}}{M} = \frac{M_{14}M_{24}}{M}\,, \end{cases} \tag{13}$$

where M denotes the total initial mass $M = M_{13} + M_{24} = M_{14} + M_{23}$.

2.5 Logarithmic Sobolev Inequality

Finally, we introduce a lemma which is known to hold, but somehow without reference. We therefore follow an argument of Strook [22], which shows that Sobolev and Poincaré inequality imply the logarithmic Sobolev inequality without confining potential on a bounded domain.

Lemma 1 (Logarithmic Sobolev inequality on bounded domains). *Let Ω be a bounded domain in $\mathbb{R}^N$ such that the Poincaré (-Wirtinger) and Sobolev inequalities*

$$\|\phi - \textstyle\int_\Omega \phi\, dx\|^2_{L^2(\Omega)} \le P(\Omega)\, \|\nabla_x \phi\|^2_{L^2(\Omega)}\,, \tag{14}$$

$$\|\phi\|^2_{L^q(\Omega)} \le C_1(\Omega)\, \|\nabla_x \phi\|^2_{L^2(\Omega)} + C_2(\Omega)\, \|\phi\|^2_{L^2(\Omega)}\,, \qquad \frac{1}{q} = \frac{1}{2} - \frac{1}{N}\,, \tag{15}$$

hold. Then, the logarithmic Sobolev inequality

$$\int_\Omega \phi^2 \log\left(\frac{\phi^2}{\|\phi\|_2^2}\right) dx \le L(\Omega,N)\, \|\nabla_x \phi\|^2_{L^2(\Omega)} \tag{16}$$

holds (for some constant $L(\Omega,N) > 0$).

Proof (of Lemma 1). Assume firstly that $\|\phi\|_2^2 = 1$. Then, using Jensen's inequality for the measure $\phi^2\, dx$, we estimate

$$\begin{aligned} \int_\Omega \phi^2 \,\log(\phi^2)\, dx &= \frac{2}{q-2}\int_\Omega \log\left(\phi^{q-2}\right)\,(\phi^2 dx) \le \frac{2}{q-2}\log\left(\int_\Omega \phi^q\, dx\right) \\ &= \frac{q}{q-2}\log\left(\|\phi\|_q^2\right) \le \frac{q}{q-2}\left(\|\phi\|_q^2 - 1\right), \end{aligned}$$

using the elementary inequality $\log x \le x - 1$. Hence, we have for general ϕ,

$$\begin{aligned} \int_\Omega \phi^2 \log\left(\frac{\phi^2}{\|\phi\|_2^2}\right) dx \le& \frac{q}{q-2}\,\left(\|\phi\|_q^2 - \|\phi\|_2^2\right) \\ \le& \frac{q}{q-2}\, C_1\, \|\nabla_x \phi\|_2^2 + \frac{q}{q-2}\,(C_2 - 1)\, \|\phi\|_2^2, \end{aligned}$$

using the Sobolev inequality (15). Now, in case when $\int_\Omega \phi \, dx = 0$, inequality (16) follows directly from Poincaré inequality (14). Otherwise, considering $\tilde{\phi} = \phi - \int_\Omega \phi \, dx$, a lengthy calculation [13] shows that

$$\int_\Omega \phi^2 \log\left(\frac{\phi^2}{\|\phi\|_2^2}\right) dx \le \int_\Omega \tilde{\phi}^2 \log\left(\frac{\tilde{\phi}^2}{\|\tilde{\phi}\|_2^2}\right) dx + 2\,\|\tilde{\phi}\|_2^2\,,$$

and the inequaltiy (16) follows from Poincaré inequality (14).

Remark 2. *On convex domains Ω, an alternative proof of* (16) *consists in building a limiting procedure with a sequence of logarithmic Sobolev inequalities on $I\!R^N$ (see e.g. [2, 6]) with a convex confining potential, which is made constant inside the bounded domain (by using the Holley-Strook perturbation lemma [19]) and tends to infinity outside of the bounded domain.*

3 Exponential Convergence to Equilibrium via the Entropy Method

In this section, we prove exponential convergence towards equilibrium (with explicit rates) for weak solutions of system (3) (and thus also for classical solution whenever they are known to exist) in all space dimensions $N \ge 1$:

Theorem 1. *Let Ω be a bounded domain with sufficiently smooth boundary (e.g. $\partial\Omega \in C^{2+\alpha}$, $\alpha > 0$) such that Lemma 1 holds. Let $(d_i)_{i=1,..,4} > 0$ be positive diffusion coefficients. Let the initial data $(a_{i,0})_{i=1,..,4}$ be nonnegative functions of $L^2\,(\log L)^2(\Omega)$ with positive masses $(M_{jk})_{(j,k)\in(\{1,2\},\{3,4\})} > 0$ (see* (4)*). Then, the global solution a_i of* (3)*–*(2) *(weak or classical as shown to exist in [7, 12]) decay exponentially towards the positive equilibrium state $(a_{i,\infty})_{i=1,..,4} > 0$ defined by* (13)*:*

$$\sum_{i=1}^{4} \|a_i(t,\cdot) - a_{i,\infty}\|^2_{L^1(\Omega)} \le C_1 \bigg(E((a_{i,0})_{i=1,..,4}) - E((a_{i,\infty})_{i=1,..,4}) \bigg)\, e^{-C_2\, t},$$

for all $t \ge 0$ and for constants C_1 and C_2, which can be explicitly computed.

Remark 3. *The above Theorem generalises to all space dimensions the convergence result obtained in [11]. It avoids a slowly growing L^∞-bound (available only in 1D and maybe 2D) by using the logarithmic Sobolev inequality* (16) *to control the relative entropy of the concentrations a_i w.r.t. their spatial averages $\overline{a_i} = \int_\Omega a_i \, dx$ (recall that $|\Omega| = 1$), which themself are controlled by the mass conservation laws* (4)*. The remaining part of the proof follows then from [11].*

Note also that exponential decay towards equilibrium in $L^p(\Omega)$ with $1 < p < 2$ follows by interpolation the $L^2(\Omega)$-bounds (12)*.*

Proof (of Theorem 1). The proof is based on an entropy method, where the entropy dissipation $D((a_i)_{i=1,..,4}) = -\frac{d}{dt}E((a_i)_{i=1,..,4}) = -\frac{d}{dt}(E((a_i)_{i=1,..,4}) - E((a_{i,\infty})_{i=1,..,4}))$ is controlled from below in terms of the relative entropy with respect to equilibrium. That is, we look for an estimate like

$$\begin{aligned} D((a_i)_{i=1,..,4}) &\geq C\,(E((a_i)_{i=1,..,4}) - E((a_{i,\infty})_{i=1,..,4})) \\ &= C\sum_{i=1}^{4}\int_{\Omega}\left[a_i \log\left(\frac{a_i}{a_{i,\infty}}\right) - (a_i - a_{i,\infty})\right] dx, \end{aligned} \tag{17}$$

for a constant C provided that all the conservation laws (4) are observed. Then, a simple Gronwall lemma yields exponential convergence in relative entropy to the equilibrium $(a_{i,\infty})_{i=1,..,4}$. Furthermore, convergence in L^1 as stated in Theorem 1 follows from a Cziszar-Kullback type inequality [11, Proposition 4.1].

In order to establish the entropy-entropy dissipation estimate (17), we firstly split the relative entropy

$$\begin{aligned} E((a_i)_{i=1,..,4}) - E((a_{i,\infty})_{i=1,..,4}) = E((a_i)_{i=1,..,4}) - E((\overline{a_i})_{i=1,..,4}) \\ + E((\overline{a_i})_{i=1,..,4}) - E((a_{i,\infty})_{i=1,..,4})\,, \end{aligned}$$

into – roughly speaking – the relative entropy of the concentrations a_i w.r.t. their averages $\overline{a_i}$ and the relative entropy of the averages $\overline{a_i}$ w.r.t. the equilibrium $a_{i,\infty}$.

The first term can be estimated thanks to the logarithmic Sobolev inequality (16) (recall the conservation laws (4)) by

$$\begin{aligned} E((a_i)_{i=1,..,4}) - E((\overline{a_i})_{i=1,..,4}) &= \sum_{i=1}^{4}\int_{\Omega} a_i \log\left(\frac{a_i}{\overline{a_i}}\right) dx \\ &\leq L(\Omega)\sum_{i=1}^{4}\int_{\Omega}\left|\nabla_x\sqrt{a_i}\right|^2 dx\,, \end{aligned}$$

which is clearly bounded by the entropy dissipation $D((a_i)_{i=1,..,4})$ in (6).

On the other hand, estimating the second relative entropy can be done in the following way: We define

$$\phi(x,y) = \frac{x\,\ln(x/y) - (x-y)}{(\sqrt{x}-\sqrt{y})^2} = \phi(x/y, 1),$$

which is a continuous function on $(0,\infty)\times(0,\infty)$. Note that thanks to the conservation laws (4), we have $\phi(\overline{a_i}/a_{i,\infty}, 1) \leq C(M)$. We can then write

$$\begin{aligned} E((\overline{a_i})_{i=1,..,4}) - E((a_{i,\infty})_{i=1,..,4}) &= \sum_{i=1}^{4}\left[\overline{a_i}\log\left(\frac{\overline{a_i}}{a_{i,\infty}}\right) - (\overline{a_i} - a_{i,\infty})\right] \\ \leq \sum_{i=1}^{4}\phi(\overline{a_i}, a_{i,\infty})\left|\sqrt{\overline{a_i}} - \sqrt{a_{i,\infty}}\right|^2 &\leq C(M)\sum_{i=1}^{4}\left|\sqrt{\overline{a_i}} - \sqrt{a_{i,\infty}}\right|^2. \end{aligned}$$

Finally, the expression $\sum_{i=1}^{4} \left|\sqrt{a_i} - \sqrt{a_{i,\infty}}\right|^2$ is bounded in terms of equation (47) in [11, Lemma 3.2], which itself is bounded by the entropy dissipation $D((a_i)_{i=1,..,4})$ in (6) with a constant, which can be explicitly estimated. This finishes the proof of the entropy entropy-dissipation estimate (17), which implies explicit exponential convergence to equilibrium in relative entropy.

The proof of Theorem 1 follows then by recalling the Cziszar-Kullback type inequality [11, Proposition 4.1].

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