Exact substitute processes for diffusion–reaction systems with local complete exclusion rules

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Abstract. Lattice systems with one species diffusion–reaction processes under local complete exclusion rules are studied analytically starting from the usual master equations with discrete variables and their corresponding representation in a Fock space. On this basis, a formulation of the transition probability as a Grassmann path integral is derived in a straightforward manner. It will be demonstrated that this Grassmann path integral is equivalent to a set of Ito stochastic differential equations. Averages of arbitrary variables and correlation functions of the underlying diffusion–reaction system can be expressed as weighted averages over all solutions of the system of stochastic differential equations. Furthermore, these differential equations are equivalent to a Fokker–Planck equation describing the probability distribution of the actual Ito solutions. This probability distribution depends on continuous variables in contrast to the original master equation, and their stochastic dynamics may be interpreted as a substitute process which is completely equivalent to the original lattice dynamics. Especially, averages and correlation functions of the continuous variables are connected to the corresponding lattice quantities by simple relations. Although the substitute process for diffusion–reaction systems with exclusion rules has some similarities to the well-known substitute process for the same system without exclusion rules, there exists a set of remarkable differences. The given approach is not only valid for the discussed single-species processes. We give sufficient arguments to show that arbitrary combinations of unimolecular and bimolecular lattice reactions under complete local exclusions may be described in terms of our approach.
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1. Introduction

In recent years, many studies have been undertaken to understand the dynamical behaviour of classical diffusive systems under exclusive restrictions [1]–[13]. This natural problem has been associated with an extraordinarily large variety of phenomena related to various chemical and diffusion processes. Usually, systems in which reactants are transported by diffusion [14, 15] are denoted as reaction–diffusion systems. Two fundamental timescales characterize these systems: (i) the diffusion time as a typical time between collisions of reacting particles and (ii) the reaction time defining the inverse reaction rate of neighbouring particles. When the reaction time is much larger than the diffusion time, the process follows approximately the classical kinetic equations. Such a process is reaction-limited. It is characterized by the validity of the law of mass action and by neglecting possible concentration fluctuations. The opposite case—diffusion-limited processes—is much more in the focus of recent investigations [16]–[21]. In this limit, local concentration fluctuations dominate the kinetics [22]–[25]. Although the effect of fluctuations is most pronounced in low dimensions or in confined geometries, there are also non-negligible effects in the traditional three-dimensional space.

The diffusion time is usually defined by the mean-square distance between reacting particles and the corresponding diffusion coefficient. Since the particle diffusion coefficient \( D \) depends on the normalized total concentration \( c \) via \( D \sim (1 - c) \), a process may also be diffusion-limited in dense systems, although the mean distance between the particles is relatively short. For instance, heterogeneous reaction systems form increasing product layers between the educts regions, so that a particle has to overcome this barrier by diffusion processes in order to find an appropriate reaction partner. It is well known that the educt-product surfaces are strongly controlled by local diffusion-induced fluctuations [26]. On the other hand, several autocatalytic reaction processes generate a transport of particle properties by chemical reactions, so that these effects also contribute to the diffusion. Consequently, the collective diffusion coefficients in dense autocatalytic systems are often controlled by the reaction time and not by the original Brownian motion.

The aim of this paper is to present a general formalism in order to describe a large class of reaction–diffusion systems obeying the exclusion principle. Although we focus our investigation on various single-species reactions, the formalism can be extended to all reaction–diffusion systems considering spontaneous changes of particles with the schematic representation \( A \rightarrow B \) as well as several pair reactions of the general type \( A + B \rightarrow C + D \).

2. Classical lattice reaction–diffusion systems

One possible way to approach diffusion and reaction processes under exclusive restrictions is to map the real dynamical processes onto a given lattice structure in the sense of a coarse-graining approximation. Usually, it means that the considered system is divided into small cells. Every cell is occupied by particles or states specified by kinetic rules depending on the situation in mind. Application of the exclusion principle means that the occupation numbers and the number of states, respectively, are restricted. The problem is, therefore, to formulate the dynamics in such a way that these restrictions are taken into account. In particular, such a situation can be described in a rather compact form starting from a master equation on a lattice [5, 27, 28].
A natural way to describe lattice-diffusion processes is given by Kawasaki exchange processes, which may be written schematically as

\[ A + 0 \rightleftharpoons 0 + A, \]  

(1)

where \( A \) represent a particle, while 0 indicates a vacancy. The elementary rule of the Kawasaki process means that the elements of a pair \((A, 0)\) exchange their positions with an empirically defined transition rate. At sufficiently large scales, the simple rule (1) leads to a diffusion equation for the continuous particle density \( \rho \)

\[ \frac{\partial \rho(\mathbf{r}, t)}{\partial t} = D \nabla^2 \rho(\mathbf{r}, t), \]  

(2)

which can be solved straightforwardly using well-known standard methods considering initial and boundary conditions. It should be remarked that the diffusion coefficient \( D \) describes the collective diffusion of the system, while the particle (or tracer-) diffusion coefficient depends on the particle concentration [29].

Additionally, we introduce lattice reactions. In this paper, we focus on single-species reactions. An important class of single-species lattice reaction processes considering the excluded volume principle are the so-called contact processes [19], defined as evolutionary rules on a set of elementary objects. Since we have only one species of particles, each site of the lattice is either occupied by a particle or is vacant. The transition rules define that a vacant site with \( n \) occupied nearest neighbours becomes also occupied with the rate \( \lambda \), while particles disappear spontaneously, i.e. independent of their surroundings, with the rate \( \beta \). These processes may be described by the reaction scheme

\[ nA + 0 \rightarrow (n + 1)A \quad \text{and} \quad A \rightarrow 0, \]  

(3)

where \( A \) represents the particles. The classical kinetic equation describing the evolution of the particle density \( \rho \) on the level of a mean-field theory at sufficiently large scales is then given by

\[ \frac{d\rho}{dt} = \lambda \rho^n (1 - \rho) - \beta \rho. \]  

(4)

This equation has two stationary solutions, the vacuum state \( \rho = 0 \) and an active phase \( \rho = \rho_0 (\lambda/\beta) > 0 \). Obviously, the vacuum state is stable in the sense that this configuration remains unchanged after the application of the evolutionary rules. Furthermore, there exist a set of initial configurations (e.g. configurations with isolated particles) belonging to the basin of attraction of the vacuum state. The active phase, characterized by a nonzero density of particles, exists only for a sufficiently large creation rate \( \lambda > \lambda_c \), otherwise the corresponding stationary solution is unstable and each initial configuration converges to the vacuum state. It should be remarked that a contact process in a \( d \)-dimensional space is associated with the directed percolation model in \( d + 1 \) dimensions since the dimension along the preferred direction of this model can be interpreted as a time axis. In this sense, the critical value \( \lambda_c \) corresponds to the directed percolation threshold.

Another important class of lattice reaction processes is given by autocatalytic processes. Here, we have a reversible reaction scheme

\[ nA + 0 \rightleftharpoons (n + 1)A. \]  

(5)
The forward reaction is again defined by the above-mentioned evolutionary rule, while the backward reaction means that a particle surrounded by \( n \) occupied nearest neighbours disappears with the rate \( \beta \). The corresponding kinetic equation now reads

\[
\frac{d\rho}{dt} = \lambda \rho^n (1 - \rho) - \beta \rho^{n+1},
\]

from which we find again two stationary solutions, the vacuum state \( \rho = 0 \) and an active phase \( \rho = \rho_0 (\lambda / \beta) = \lambda / (\lambda + \beta) > 0 \). The vacuum state is again stable in the above-mentioned sense, but its basin of attraction contains only the vacuum state itself. There are other stable configurations (e.g. in case of \( n \geq 2 \) configurations with only isolated particles) that are not considered in the classical solution, but the probability that such an initial configuration can be obtained by a certain random process generating all allowed configurations converges to zero for an infinitely large lattice. Thus, the mean field solution \( \rho = 0 \) is unstable, while the corresponding solution \( \rho_0 \) for the active phase is always stable.

Both contact and autocatalytic processes may be extended by the Kawasaki exchange process (1) in order to introduce diffusion rules. Together with the chemical reaction equations, we arrive now at the continuous mean-field equation for the particle concentration

\[
\frac{\partial \rho \left( \mathbf{r}, t \right)}{\partial t} = D \nabla^2 \rho \left( \mathbf{r}, t \right) + a \rho \left( \mathbf{r}, t \right) - \sum_{k \geq 2} b_k \rho^k \left( \mathbf{r}, t \right).
\]

The coefficients \( a \) and \( b_k \) depend on the underlying chemical process and the kinetic order \( n \). It should be remarked that, in the case of a spatially homogeneous system \( \rho \left( \mathbf{r}, t \right) = \rho \left( t \right) \), we find again the kinetic equations (4) and (6). As mentioned above, we will restrict our investigation to the analysis of pair reactions and spontaneous decays. This means that we have to deal with autocatalytic and contact processes of order \( n = 1 \). Thus, we get

\[
a = \lambda - \beta \quad \text{and} \quad b_2 = \lambda
\]

in case of the contact process, while the autocatalytic process requires

\[
a = \lambda \quad \text{and} \quad b_2 = \lambda + \beta.
\]

Also in the presence of diffusion processes, we find an active phase for \( a, b > 0 \). However, the kinetic equations (4), (6) and (7) consider only the mean evolution from an initial state far from the stationary state into the stationary state. Close to the stationary state, fluctuations due to the random realization of the given rules become dominant. In order to keep the influence of these fluctuations, (7) has to be modified by introducing a noise term [30]

\[
\frac{\partial \rho \left( \mathbf{r}, t \right)}{\partial t} = D \nabla^2 \rho \left( \mathbf{r}, t \right) + a \rho \left( \mathbf{r}, t \right) - \sum_{k \geq 2} b_k \rho^k \left( \mathbf{r}, t \right) + \sqrt{\rho \left( \mathbf{r}, t \right)} \eta \left( \mathbf{r}, t \right).
\]

It seems surprising that the noise is multiplicatively coupled, but a detailed mathematical analysis [31, 32] shows that this square root is the proper term under the customary assumption of Poissonian statistics for the reaction part. It should be remarked that there is an additional noise due to the random realization of the diffusive process. But this noise does not affect the critical
behaviour of (10) so that we can neglect it for the moment. The noise \( \eta(r, t) \) has a Gaussian character, i.e. we obtain

\[
\langle \eta(r, t)\eta(r', t') \rangle \sim \delta(r - r')\delta(t - t').
\]

When these fluctuations are taken into account, the critical point separating the vacuum state from the active phase shifts from \( a = 0 \) to \( a_c > 0 \) and the critical behaviour is nonclassical. That is, the autocatalytic process now also has a stable vacuum state corresponding to \( \lambda < a_c \), while the contact process requires \( \lambda < a_c + \beta \) for a stable vacuum state. Furthermore, the nonclassical critical behaviour is characterized by new scaling exponents. For example, the mean density scales now as \( \rho \sim (a - a_c)^\zeta \), with \( \zeta = 0.277 \) in \( d = 1 \), while the mean field exponent is given by \( \zeta = 1 \).

Obviously, the critical behaviour will not be changed if we try to consider the excluded volume effect properly in the analytical description of these diffusion–reaction processes. We remark that the stochastic equation (10) is a coarse-grained evolution equation considering lattice rules only in the collective diffusion coefficient and reaction rates and in the coupling with the noise term. Especially, the exclusion principle itself was not considered by the derivation of (10). Therefore, it may be expected that the evolution of the density at short and moderate times and length scales of a real lattice model is significantly affected by the presence of this principle, so that, in this case, (10) must be replaced by a more general equation.

The aim of the present paper is to obtain a microscopic representation of autocatalytic or contact single-species diffusion–reaction processes of kinetic order \( n = 1 \) under a proper consideration of the mutual exclusions in terms of a generalized Fokker–Planck equation. We expect that the microscopic representation with exclusion principle approaches the briefly discussed classical kinetic equation (4), (6), (7) and (10) at large distances and for long times, while significant differences should remain for short and moderate scales.

3. Path integral representation

3.1. Master equations and the Fock-space approach

3.1.1. General remarks. We start our investigation from the usual master equations and the corresponding representation in Fock space, respectively. The original master equation contains discrete observables describing the actual occupation of the lattice sites. For the processes in mind, the occupation of a lattice site \( i \) is determined by a number \( n_i \) with \( n_i = 1 \) if a particle \( A \) occupies the lattice site and \( n_i = 0 \) if the site is vacant. The set \( n = \{n_j\} \) of all occupation numbers forms a configuration. From this point of view, the evolution of the statistical probability distribution function \( P(n, t) \) can be described by a master equation:

\[
\frac{\partial P(n, t)}{\partial t} = \sum_{n'} L(n, n') P(n', t),
\]

where the dynamical matrix \( L(n, n') \) contains all information about the kinetic rules of the lattice system.

Following Doi [27] (compare also [5]), the probability distribution \( P(n, t) \) can be related to a state vector \( |F(t)\rangle \) in a Fock space according to \( P(n, t) = \langle n | F(t) \rangle \) and therefore
$|F(t)⟩ = \sum_n P(n, t)|n⟩$, respectively, with basis vectors $|n⟩$. Using this representation, the master equation (12) can be transformed to an equivalent equation in the Fock space

$$\partial_t |F(t)⟩ = \hat{L}|F(t)⟩.$$  \hspace{1cm} (13)

The dynamical matrix $L(n, n')$ of (12) is now mapped onto the evolution operator

$$\hat{L} = \sum_{n, n'} |n⟩L(n, n')⟨n'|.$$  \hspace{1cm} (14)

Usually, this operator is expressed in terms of creation and annihilation operators with Bose properties [27, 28, 33] or Pauli properties [2, 5, 8, 28]. In order to derive a path integral formulation considering the exclusion principle, we have to construct the evolution operator on the basis of Fermi operators.

### 3.1.2. Evolution operator for local spontaneous reactions

First of all, we investigate the spontaneous creation $0 \rightarrow A$ and annihilation $A \rightarrow 0$ of a particle at a certain lattice site with the creation rate $\lambda$ and the annihilation rate $\beta$. The master equations of these processes can be written as

$$\partial_t P(A) = \lambda P(0) - \beta P(A) \quad \text{and} \quad \partial_t P(0) = \beta P(A) - \lambda P(0).$$  \hspace{1cm} (15)

In principle, two basis states are sufficient for the representation of this problem using the Fock-space method. In order to transfer the exclusion principle (the lattice site is either occupied by only one particle or it is vacant) onto the algebraic properties of some basis operators, it seems reasonable to use a Clifford (or Fermi-) algebra, which is characterized by characteristic anticommutation relations. But this approach contains an intrinsic problem. Later we are interested in the extension of the simple one-site problem to a complete lattice problem. This extension implies serious difficulties related to the commutation rules of operators and states. Especially, such a method requires unwanted and uncomfortable manipulations of the sign of the lattice state. Thus, the standard procedure to is apply a Pauli algebra, which is Fermi-like for operators acting at the same lattice site, but Bose-like for operators at different sites [34, 35]. Unfortunately, this algebra is not suitable for the generation of clear path integrals. Therefore, we will choose an alternative approach. To achieve this aim, we introduce a Fock space $\mathcal{F}$ defined by the four basis states:

$$|n^+, n^-⟩ = |n^+⟩|n^-⟩ \quad \text{with} \quad n^+, n^- \in (0, 1).$$  \hspace{1cm} (16)

$|n^+ = 1⟩ (|n^+ = 0⟩)$ describes the situation that at the lattice site particle A is present (absent). Analogously, $|n^- = 1⟩ (|n^- = 0⟩)$ is the state with the vacancy present (absent) at this lattice site. This construction, although complicated at the first sight, is necessary for the reasons given above. The algebra is completed with the introduction of the usual Fermi operators $\hat{a}_1$, $\hat{a}^\dagger_1$, $\hat{a}_2$ and $\hat{a}^\dagger_2$. These operators form a Clifford algebra with the well-known anticommutation relations:

$$\{\hat{a}_a, \hat{a}^\dagger_b\} = \delta_{ab}.$$  \hspace{1cm} (17)
\[ \{ \hat{a}_a, \hat{a}_b \} = \{ \hat{a}_a^\dagger, \hat{a}_b^\dagger \} = 0. \tag{18} \]

The application of these operators onto the basis vectors \( |n \rangle \) leads to

\[ \hat{a}_1| (n^+, n^-) \rangle = n^+| (n^+ - 1, n^-) \rangle, \quad \hat{a}_1^\dagger| (n^+, n^-) \rangle = (1 - n^+)| (n^+ + 1, n^-) \rangle \tag{19} \]

and

\[ \hat{a}_2| (n^+, n^-) \rangle = n^-(-1)^n^+| (n^+, n^- - 1) \rangle, \quad \hat{a}_2^\dagger| (n^+, n^-) \rangle = (1 - n^-)(-1)^n^+| (n^+, n^- + 1) \rangle. \tag{20} \]

Of course, the dimension of the chosen Fock space is too large for the description of the two allowed states. In order to represent the presence or absence of the particle A (and therefore the absence or presence of a vacancy), we need only two of the four available Fock-space basis states. It seems reasonable to choose the identification

\[ A \rightarrow |(1, 0) \rangle = |1 \rangle \quad \text{and} \quad 0 \rightarrow |(0, 1) \rangle = |0 \rangle. \tag{21} \]

The two states form the subspace \( \mathcal{F}_s \) of the total Fock space \( \mathcal{F} (\mathcal{F}_s \subset \mathcal{F}) \). Obviously, the two basis states \( |n \rangle = |0 \rangle, |1 \rangle \) of this subspace satisfy the condition

\[ (\hat{a}_1^\dagger \hat{a}_2 + \hat{a}_2^\dagger \hat{a}_1) |n \rangle = |n \rangle, \tag{22} \]

while the other two basis states of the complete Fock space violate this relation. After fixing the basis states, we can represent the master equation (15) in terms of this reduced basis. It is clear that the application of the evolution operator \( \hat{L} \) should not lead to a state outside the subspace \( \mathcal{F}_s \). In other words, each possible term of the evolution operator is a combination of creation and annihilation operators such that its application onto an arbitrary basis state (21) \( |n \rangle = |(n^+, n^-) \rangle \in \mathcal{F}_s \) guarantees the conservation \( n^+ + n^- = 1 \). It can be checked that each operator of the form

\[ \hat{L} = K_{12} \hat{a}_1^\dagger \hat{a}_2 + K_{21} \hat{a}_2^\dagger \hat{a}_1 + K_{11} \hat{a}_1^\dagger \hat{a}_1 + K_{22} \hat{a}_2^\dagger \hat{a}_2 \tag{23} \]

fulfils this condition. Furthermore, the configuration state \( |F(t) \rangle \) is defined by

\[ |F(t) \rangle = P(A, t)|1 \rangle + P(0, t)|0 \rangle. \tag{24} \]

The master equations (15) can be obtained directly from the general evolution equation \( \partial_t |F(t) \rangle = \hat{L} |F(t) \rangle \) due to the orthogonality of the basis states \( \langle n'|n \rangle = \delta_{n'}^n \) when we identify \( K_{11} = -\beta, K_{12} = \lambda, K_{21} = \beta \) and \( K_{22} = -\lambda \). Thus, (13) together with (23) and (24) can be interpreted as the Fock space representation of the master equations (15).

### 3.1.3. Spontaneous lattice reactions

The Fock space \( \mathcal{F} \) (and therefore also the reduced Fock space \( \mathcal{F}_s \)) of a spontaneous single-species reaction at a certain site can be extended to a Fock space (and therefore to the corresponding reduced Fock space) of independent spontaneous single-species reactions on \( N \) lattice sites. The basis states of the reduced Fock space \( \mathcal{F}_s \) are now
defined by the ordered product of local basis states

$$|n⟩ = \prod_{i=1}^{N} |n_i⟩ = |n_1⟩|n_2⟩ \cdots |n_N⟩.$$  \hspace{1cm} (25)

The $N$ factors of the vector $n$ are the local occupation numbers $n_i \in (0, 1)$ corresponding to the states (21) of the reduced Fock space. The state $|F(t)⟩$ is again a linear combination of all basis vectors

$$|F(t)⟩ = \sum_n P(n, t) |n⟩,$$

and the evolution operator becomes

$$\hat{L} = \sum_{i=1}^{N} \hat{L}_i \quad \text{with} \quad \hat{L}_i = \lambda \hat{a}_1^\dagger \hat{a}_2,i + \beta \hat{a}_2^\dagger \hat{a}_1,i - \beta \hat{a}_1^\dagger \hat{a}_1,i - \lambda \hat{a}_2^\dagger \hat{a}_2,i$$

(27)

because of the identity and the physical independence of the local spontaneous reactions. Here, we may see the consequence of our choice of the basis states (21). The action of an arbitrary local term of the evolution operator related to the position $i$ changes the state $|n⟩$ at this position, i.e. $|n_1⟩ \cdots |n_i⟩ \cdots |n_N⟩ \rightarrow |n_1⟩ |n_1'⟩ \cdots |n_N⟩$, but the sign of the state remains unchanged due to the bilinear structure of all terms and the special structure of the basis states of the reduced Fock space. In the case of the traditional Fermionic Fock space, the evolution operator contains also linear terms describing the creation and annihilation of particles, so that now we have to deal with position-dependent signs of the total basis states after each application $\hat{L}$. Finally, it should be remarked again that the anticommutation relations are completely defined by the well-known Fermionic algebra:

$$\{ \hat{a}_{a,i}, \hat{a}_{b,j}^\dagger \} = \delta_{ab} \delta_{ij} \quad \{ \hat{a}_{a,i}, \hat{a}_{b,j} \} = \{ \hat{a}_{a,i}^\dagger, \hat{a}_{b,j}^\dagger \} = 0.$$  \hspace{1cm} (28)

3.1.4. Kinetic coupling: autocatalytic reactions. The kinetic restrictions of an elementary autocatalytic step allow the realization of an elementary change $A \leftrightarrow 0$ at the lattice site $i$ only if at least one neighbour site is also occupied. This restriction can be considered in the evolution operator $\hat{L}$ by the following extension:

$$\hat{L} = \sum_{i,j} \Theta_{ij} \hat{a}_{1,j}^\dagger \hat{a}_{1,i} \hat{L}_i$$

(29)

($\Theta_{ij}$ is a lattice indicator with $\Theta_{ij} = 1$ if $i$ and $j$ are neighbouring sites, otherwise $\Theta_{ij} = 0$). The additional factor $\hat{a}_{1,j}^\dagger \hat{a}_{1,i}$ allows a change of the lattice configuration by a simple unimolecular reaction $A \rightarrow 0$ at the lattice site $i$ if the neighbouring site $j$ is occupied, i.e. if $|n_j⟩ = |1⟩$. Otherwise, the application of $\hat{a}_{1,j}^\dagger \hat{a}_{1,i}$ onto the state $|n_j⟩ = |0⟩$ leads to $\hat{a}_{1,j}^\dagger \hat{a}_{1,i} |0⟩ = 0$. Thus the evolution operator of the autocatalytic reactions may be explicitly written as

$$\hat{L}_{\text{auto}} = \sum_{i,j} \Theta_{ij} \hat{a}_{1,j}^\dagger \hat{a}_{1,i} \left[ \lambda \hat{a}_{2,i}^\dagger \hat{a}_{2,i} + \beta \hat{a}_{2,i}^\dagger \hat{a}_{1,i} - \beta \hat{a}_{1,i}^\dagger \hat{a}_{1,i} - \lambda \hat{a}_{2,i}^\dagger \hat{a}_{2,i} \right]$$

(30)
with the rate $\lambda$ for the forward reaction $A + 0 \rightarrow 2A$ and the rate $\beta$ for the backward reaction $2A \rightarrow A + 0$. Furthermore, the contact process may be interpreted as the superposition of autocatalytic forward reactions with the rate $\lambda'$ and a spontaneous decay $A \rightarrow 0$ with the rate $\beta'$. We get

$$\hat{L}_{\text{contact}} = \lambda' \sum_{i,j} \Theta_{ij} \hat{a}_{1,j}^\dagger \hat{a}_{1,j} \left[ \hat{a}_{2,i}^\dagger \hat{a}_{2,i}^\dagger - \hat{a}_{2,i}^\dagger \right] + \beta' \sum_{i} \left[ \hat{a}_{2,i}^\dagger \hat{a}_{1,i}^\dagger - \hat{a}_{1,i}^\dagger \right]. \quad (31)$$

The evolution operators (30) and (31) may be combined to a general operator describing both autocatalytic and contact processes

$$\hat{L}_{\text{chem}} = \widetilde{\lambda} \sum_{i,j} \Theta_{ij} \hat{a}_{1,j}^\dagger \hat{a}_{1,j} \left( \hat{a}_{1,i}^\dagger - \hat{a}_{2,i}^\dagger \right) \hat{a}_{2,i} + \sum_{i,j} \Theta_{ij} \left[ \beta \hat{a}_{1,j}^\dagger \hat{a}_{1,j} + \frac{\beta'}{z} \right] \left( \hat{a}_{2,i}^\dagger - \hat{a}_{1,i}^\dagger \right) \hat{a}_{1,i} \quad (32)$$

with $\widetilde{\lambda} = \lambda + \lambda'$ and $z$ the coordination number of the lattice.

3.1.5. Diffusion processes. Following the above-discussed construction principle for the evolution operator, Kawasaki exchange processes $A + 0 \rightleftharpoons 0 + A$ are described by

$$\hat{L}_{\text{diff}} = \frac{D}{2} \sum_{i,j} \Theta_{ij} \left[ \hat{a}_{2,i}^\dagger \hat{a}_{1,i}^\dagger \hat{a}_{1,i} \hat{a}_{2,i} + \hat{a}_{1,i}^\dagger \hat{a}_{2,i}^\dagger \hat{a}_{2,i}^\dagger \hat{a}_{1,i}^\dagger - \hat{a}_{2,i}^\dagger \hat{a}_{1,i}^\dagger \hat{a}_{1,i} \hat{a}_{2,i} - \hat{a}_{1,i}^\dagger \hat{a}_{2,i}^\dagger \hat{a}_{2,i} \hat{a}_{1,i}^\dagger \right], \quad (33)$$

where we have introduced the diffusion coefficient $D$ controlling the jump rate of a particle $A$ between neighbouring sites. The superposition of the evolution operators for the diffusion steps (33) and for the chemical reaction processes (32) leads to the total evolution operator of the diffusion–reaction system with exclusion, $\hat{L} = \hat{L}_{\text{diff}} + \hat{L}_{\text{chem}}$.

It should be remarked that the presented Fock-space formalism has the decisive advantage of a simple construction principle for each evolution operator $\hat{L}$ on the basis of creation and annihilation operators. Therefore, this method allows investigations of master equations for various evolution processes, e.g. aggregation, chemical reactions [34, 35], nonthermal kinetic Ising models [36], nonlinear diffusion [37] and just reaction–diffusion models.

3.2. General properties

3.2.1. Normalization condition. Using the representation (26) and the orthogonality of the basis states, one obtains the normalization condition which must hold for all master equations

$$\sum_{m} \langle m | F(t) \rangle = \sum_{n,m} P(n, t) \langle m | n \rangle = \sum_{m} P(m, t) = 1. \quad (34)$$

This important property can be simply written as $\langle s | F(t) \rangle \equiv 1$ by using the so-called reference state $| s \rangle$.

$$\langle s | = \sum_{m} \langle m |. \quad (35)$$

From this point of view, one obtains from (13) a second possibility to describe the normalization condition

$$\partial_{t} \langle s | F(t) \rangle = \langle s | \hat{L} | F(t) \rangle = 0 \Rightarrow \langle s | \hat{L} = 0. \quad (36)$$

Hence, an evolution operator $\hat{L}$ conserves the total probability if and only if $\langle s | \hat{L} = 0$. 

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3.2.2. Averages and operators. The reference state (35) can be used also for the representation of averages. The average of a given observable $C(n)$ as a function of the actual occupation $n$ is usually defined by $\overline{C} = \sum_n P(n, t) C(n)$. On the other hand, the observable $C(n)$ can be associated with an operator $\hat{C} = \sum_n |n\rangle C(n) \langle n|$. Thus, using (26), the average $\overline{C}$ can be simply written as

$$\overline{C}(t) = \langle s | \hat{C} | F(t) \rangle.$$  (37)

The average $\overline{C}(t)$ is time-dependent except that $|F(t)\rangle$ represents the equilibrium or another stationary state. Furthermore, a formal equation of motion for this average follows from (37) and (13):

$$\partial_t \overline{C}(t) = \langle s | \hat{C} \hat{L} | F(t) \rangle.$$  (38)

For the investigations below, we need the following two operators:

$$\hat{A}^+ = \hat{a}^{\dagger}_{1,i} \hat{a}_{1,i} \quad \text{and} \quad \hat{A}^- = \hat{a}^{\dagger}_{2,i} \hat{a}_{2,i}.$$  (39)

These operators indicate the occupation of a given lattice site $i$, i.e. $\hat{A}^+$ corresponds to an occupied site, while $\hat{A}^-$ is related to a vacancy. Additionally, $\hat{A}^+$ and $\hat{A}^-$ allow the projection of the reference state $\langle s \rangle$ onto an arbitrary basis state representing the configuration $n$ via

$$\langle n \rangle = \langle s \prod_i \left[ (\hat{A}^+)^n (\hat{A}^-)^{1-n} \right].$$  (40)

Thus, the probability $P(n, t) = \langle n | F(t) \rangle$ can be interpreted as the average of the operator $\prod_i (\hat{A}^+)^n (\hat{A}^-)^{1-n}$.

On the other hand, the average $\overline{A}^+_i = \langle s | \hat{A}^+_i | F(t) \rangle$ is equivalent to the occupation probability of the lattice site $i$, while $\overline{A}^-_i$ is the probability that the lattice site $i$ is vacant. From this point of view, $\overline{A}^+_i$ can be identified as the averaged concentration $c_i$ of particles at site $i$, i.e. $\overline{A}^+_i = c_i$, and therefore $\overline{A}^-_i = 1 - c_i$.

The operators $\hat{A}^+_i$ and $\hat{A}^-_i$ fulfil the following identities in the reduced Fock space $\mathcal{F}_i$:

$$\hat{A}^+_i \hat{A}^-_i \equiv 0 \quad \text{and} \quad \hat{A}^+_i + \hat{A}^-_i \equiv 1.$$  (41)

These identities can be used for a change of operator structures, e.g. the operators $\hat{C}$ and $\hat{C}'$ obtained from the mapping

$$\hat{C} \rightarrow \hat{C}' = \hat{C} + \gamma_1 \hat{A}^+_i \hat{A}^-_i + \gamma_2 (\hat{A}^+_i + \hat{A}^-_i - 1)$$  (42)

are identical with respect to the reduced Fock space.

3.2.3. Transition probability. The formal solution of the evolution equation (13) is given by

$$|F(t)\rangle = \exp(\hat{L}t) |F(0)\rangle.$$  (43)
Using (26), the probability distribution $P(n, t)$ can be written as

$$P(n, t) = \langle n | F(t) | m \rangle = \sum_m \langle n | \exp(\hat{L}t) | m \rangle P(m, 0). \quad (44)$$

On the other hand, the evolution of the probability function $P(n, t)$ is given by the well-known Chapman–Kolmogorov equation [40], i.e.

$$P(n, t) = \sum_m K(n, t; m, 0) P(m, 0) \quad (45)$$

with the transition probability $K(n, t; m, 0)$ describing the transition from the configuration $m$ at the initial time $t = 0$ to the configuration $n$ at time $t$. The comparison between (44) and (45) leads to the following expression for the transition probability:

$$K(n, t; m, 0) = \langle n | \exp(\hat{L}t) | m \rangle. \quad (46)$$

### 3.2.4. Correlation functions.

The correlation between two observables $C(n)$ and $D(m)$ is usually defined by

$$\overline{C(t)D(t')} = \sum_{n,m} C(n)K(n, t; m, t')D(m)P(m, 0). \quad (47)$$

A simple calculation using $\hat{C} = \sum_n |n\rangle C(n)\langle n|$ and $\hat{D} = \sum_n |n\rangle D(n)\langle n|$ and the orthogonality of the basis states, i.e. $\langle k|n\rangle = \delta_{k,n}$, yields

$$\overline{C(t)D(t')} = \sum_{n,m,k,l} \langle k|\hat{C}\exp(\hat{L}(t-t'))\hat{D}|l\rangle P(l, t')$$

$$= \sum_{k,l} \langle s|\hat{C}\exp(\hat{L}(t-t'))\hat{D}|F(t')\rangle.$$

(48)

As mentioned above, the Fock space representation is an elegant method especially for describing relatively complex master equations in terms of a quantum mechanical language. Within this framework, further calculations, e.g. different kinds of mean-field approaches or perturbation theory, are always possible [5, 34, 35, 37].

### 3.3. Grassmann path integral

The transition from a Clifford algebra (defined by the anticommutation rules of the operators $\hat{a}^\dagger_{a,i}$ and $\hat{a}_{a,i}$) to a Grassmann algebra (given by the Grassmann variables $\overline{a}_{a,i}$) is possible because of the isomorphism [38]

$$\hat{a}^\dagger_{a,i} \rightarrow \overline{a}_{a,i}, \quad \hat{a}_{a,i} \rightarrow \frac{\partial}{\partial \overline{a}_{a,i}}. \quad (49)$$

The knowledge of the evolution operator $\hat{L}$ in (13), see for example (29), allows the derivation of the transition probability $K(n, t; m, 0)$ (46) as Grassmann path integral. We use a representation
with a discrete time lattice with the sites $t_m = m(t/M)$ and the elementary time interval $\Delta t = t/M$. $m$ is an integer with $0 \leq m \leq M$. The transition probability $K(n, t; m, 0)$ follows after carrying out the limit $M \to \infty$. Using the Grassmann variables $\vec{\alpha} = (\alpha_{1,1}, \alpha_{2,1}, \ldots, \alpha_{N,1}, \alpha_{2,N})$ and $\alpha = (\alpha_{1,1}, \alpha_{1,2}, \alpha_{2,2}, \ldots, \alpha_{1,N}, \alpha_{2,N})$, where $N$ is the number of lattice sites, one obtains [39]

$$K(n, t; m, 0) = \lim_{M \to \infty} \int \left[ \prod_{a=1}^2 \prod_{i=1}^N \prod_{m=0}^{M-1} (d\alpha_a(t_m) d\alpha_a(t_m)) \right] \langle n | \exp[-\alpha(t_m) \alpha(t_m)] \rangle \\
\times \prod_{i,j} \exp \left\{ \sum_{m=0}^{M-1} \mathcal{L}_{ij}(\alpha(t_m+1), \alpha(t_m)) \Delta t \right\} \prod_{i} \exp \left\{ \sum_{m=0}^{M-1} \mathcal{L}_i \left( \alpha(t_m), \alpha(t_m) \right) \Delta t \right\} \\
\times \exp \left\{ \sum_{m=0}^{M-1} [\alpha(t_{m+1}) - \alpha(t_m)] \alpha(t_m) \right\} |n\rangle'. \hspace{1cm} (50)$$

The basis states $|n\rangle'$ and $|n\rangle$ defining the initial and final configurations $n'$ and $n$, respectively, are now ordered products of the completely anticommutating Grassmann variables:

$$|n\rangle' = \prod_i \left[ (\alpha_{1,i}(0))^{n_i} (\alpha_{2,i}(0))^{1-n_i} \right] \quad \text{and} \quad |n\rangle = \prod_i \left[ (\alpha_{2,i}(t_M))^{1-n_i} (\alpha_{1,i}(t_M))^{n_i} \right]. \hspace{1cm} (51)$$

The functions $\mathcal{L}_{ij}$ and $\mathcal{L}_i$ are obtained from the original evolution operator $\hat{L} = \hat{L}_{\text{diff}} + \hat{L}_{\text{chem}}$, with (33) and (32). Using the usual rules [38, 39] for the transition from Fermi operators to Grassmann variables, one obtains

$$\mathcal{L}_i(\vec{\alpha}, \alpha) = \beta^j(\vec{\alpha}_{2,i} - \vec{\alpha}_{1,i}) \alpha_{1,i} \hspace{1cm} (52)$$

and

$$\mathcal{L}_{ij}(\vec{\alpha}, \alpha) = \Theta_{ij} \left[ \lambda(\vec{\alpha}_{1,i} \alpha_{2,j} - \vec{\alpha}_{2,i} \alpha_{1,j}) \alpha_{1,i} \alpha_{1,j} + \beta(\vec{\alpha}_{2,i} \alpha_{1,j} - \vec{\alpha}_{1,i} \alpha_{1,j}) \alpha_{1,i} \alpha_{1,j} \right] \hspace{1cm} (53)$$

The first exponential factor in the 2nd line of (50) can be separated into a product:

$$\exp\{\mathcal{L}_{ij}(\vec{\alpha}, \alpha) \Delta t\} = \exp\{\Theta_{ij}\lambda(\vec{\alpha}_{1,i} \alpha_{2,j} - \vec{\alpha}_{2,i} \alpha_{1,j}) \alpha_{1,i} \alpha_{1,j} \Delta t\} \\
\times \exp\{\Theta_{ij}\beta(\vec{\alpha}_{2,i} \alpha_{1,j} - \vec{\alpha}_{1,i} \alpha_{1,j}) \alpha_{1,i} \alpha_{1,j} \Delta t\} \\
\times \exp\{\Theta_{ij} D(\vec{\alpha}_{2,i} \alpha_{1,j} - \vec{\alpha}_{1,i} \alpha_{1,j}) \alpha_{2,i} \alpha_{1,j} \Delta t\} \\
\times \exp\{-\Theta_{ij} D(\vec{\alpha}_{1,i} \alpha_{2,j} - \vec{\alpha}_{2,i} \alpha_{1,j}) \alpha_{1,i} \alpha_{1,j} \Delta t\}. \hspace{1cm} (54)$$

Now we consider the first factor of (54). Taking into account the complete anticommutation rules of the Grassmann variables, one obtains for $i \neq j$:

$$r_{ij}^{(1)} = \lambda(\vec{\alpha}_{1,i} \alpha_{2,j} - \vec{\alpha}_{2,i} \alpha_{1,j}) \alpha_{1,i} \alpha_{1,j} = \frac{1}{2} \left[ \sqrt{\lambda(\vec{\alpha}_{1,i} \alpha_{1,j} + (\vec{\alpha}_{1,i} - \vec{\alpha}_{2,i}) \alpha_{2,i})} \right]^2. \hspace{1cm} (55)$$

The introduction of ghost variables $\varphi_{ij}$ leads to a quadratic representation

$$\exp\{\Theta_{ij} r_{ij}^{(1)} \Delta t\} = \exp \left\{ \frac{\Delta t}{2} \left[ \Theta_{ij} \sqrt{\lambda(\vec{\alpha}_{1,i} \alpha_{1,j} + (\vec{\alpha}_{1,i} - \vec{\alpha}_{2,i}) \alpha_{2,i})} \right]^2 \right\} \\
= \mathcal{N} \int d\varphi_{ij} \exp \left\{ \sqrt{\lambda} \Delta t \Theta_{ij} \varphi_{ij} (\vec{\alpha}_{1,i} \alpha_{1,j} + (\vec{\alpha}_{1,i} - \vec{\alpha}_{2,i}) \alpha_{2,i}) \right\} \exp \left\{ -\frac{\Delta t}{2} \varphi_{ij}^2 \right\}. \hspace{1cm} (56)$$

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The normalization factor $N$ is irrelevant for the further treatment, therefore we continue without an explicit representation of this quantity. The second factor of (54) is rewritten by analogous steps and one obtains

$$
\beta(\alpha_2,\alpha_1)\alpha_1 = \frac{1}{2}[\sqrt{\beta}(\alpha_1 + (\alpha_2 - \alpha_1))]^2.
$$

(57)

Introducing the ghost variables $\psi_{ij}$, we arrive at

$$
\exp{\{\Theta_{ij}r_{ij}^{(2)}\Delta t\}} = N \int d\psi_{ij} \exp\left\{\sqrt{\beta}\Delta t\psi_{ij}(\alpha_1 + \alpha_2 - \alpha_1)\right\} \exp\left\{-\frac{\Delta t}{2}\psi_{ij}^2\right\}.
$$

(58)

Finally, the third term becomes

$$
r_{ij}^{(3)} = D\alpha_2,\alpha_1 = \frac{1}{2}[\sqrt{D}(\alpha_2 + \alpha_1)]^2
$$

and therefore

$$
\exp{\{\Theta_{ij}r_{ij}^{(3)}\Delta t\}} = N \int d\omega_{ij} \exp\left\{\sqrt{D}\Delta t\omega_{ij}(\alpha_1 + \alpha_2 - \alpha_1)\right\} \exp\left\{-\frac{\Delta t}{2}\omega_{ij}^2\right\}
$$

(60)

and the last term

$$
r_{ij}^{(4)} = -D\alpha_2,\alpha_1 = \frac{1}{2}[\sqrt{D}(\alpha_2 - \alpha_1)]^2
$$

leads to

$$
\exp{\{-\Theta_{ij}r_{ij}^{(4)}\Delta t\}} = N \int d\tilde{\omega}_{ij} \exp\left\{\sqrt{D}\Delta t\tilde{\omega}_{ij}(\alpha_1 - \alpha_1)\right\} \exp\left\{-\frac{\Delta t}{2}\tilde{\omega}_{ij}^2\right\}.
$$

(62)

At this point, it should be remarked that only the apparently complicated Fermionic structure of the problem with operators for particles and vacancies allows the manipulations above. The fact that a reaction or diffusion step requires the annihilation of the old state and the creation of the new state generates, for all allowed elementary steps, an evolution operator that consists of the same number of creation and annihilation operators. Especially, this remarkable property allows us to use operators with a Fermionic algebra for a consistent formulation of the master equation in the Fock space representation and, furthermore, to introduce a Grassmann path integral.

Another important remark is that an analogous transformation for three-particle processes (and also higher processes) leads to quadratic Grassmann terms containing nonlinear contributions of the ghost variables. These nonlinearities prevent, for three-particle and higher processes, a successful execution of the concept suggested below. Only two-particle processes lead always to a linear coupling between the quadratic Grassmann terms and the ghost variables. Fortunately, many higher-order reactions can be decomposed in a sequence of successive unimolecular and bimolecular reactions, so that the present technique is also available for these chemical processes.
The path integral (50) can now be rewritten. Using (56), (58), (60) and (62), we get

\[
K(n, t; n', 0) = \tilde{N} \lim_{M \to \infty} \int \prod_{m=1}^{M} \prod_{i,j} (\mathcal{D} \varphi_i^m \mathcal{D} \psi_j^m \mathcal{D} \omega_i^m \mathcal{D} \tilde{\omega}_j^m) \\
\times \exp \left\{ -\frac{\Delta t}{2} \sum_{m=1}^{M} \sum_{i,j} \left[ (\varphi_i^m)^2 + (\psi_j^m)^2 + (\omega_i^m)^2 + (\tilde{\omega}_j^m)^2 \right] \right\} \\
\times \int \left[ \prod_{a=1}^{2} \prod_{i=1}^{N} \prod_{m=0}^{M} (\mathcal{D} \alpha_{a,i}^m \mathcal{D} \alpha_{i,a}^m) \right] \langle n | \exp \{ -\alpha^M \} \rangle \mathcal{D} \psi_{ij}(t) \\
\times \exp \left\{ \sqrt{\lambda} \Delta t \sum_{m=0}^{M-1} \sum_{i,j} \Theta_{ij} \varphi_{ij}^{m+1} (\alpha_{i,j}^{m+1} \alpha_{i,j}^m + \alpha_{i,j}^{m+1} \alpha_{i,j}^m - \alpha_{i,j}^{m+1} \alpha_{i,j}^m) \right\} \\
\times \exp \left\{ \sqrt{\beta} \Delta t \sum_{m=0}^{M-1} \sum_{i,j} \Theta_{ij} \varphi_{ij}^{m+1} (\alpha_{i,j}^{m+1} \alpha_{i,j}^m + \alpha_{i,j}^{m+1} \alpha_{i,j}^m - \alpha_{i,j}^{m+1} \alpha_{i,j}^m) \right\} \\
\times \exp \left\{ \sqrt{D} \Delta t \sum_{m=0}^{M-1} \sum_{i,j} \Theta_{ij} \alpha_{ij}^{m+1} (\alpha_{i,j}^{m+1} \alpha_{i,j}^m + \alpha_{i,j}^{m+1} \alpha_{i,j}^m - \alpha_{i,j}^{m+1} \alpha_{i,j}^m) \right\} \\
\times \exp \left\{ \sqrt{D} \Delta t \sum_{m=0}^{M-1} \sum_{i,j} \Theta_{ij} \tilde{\alpha}_{ij}^{m+1} (\tilde{\alpha}_{i,j}^{m+1} \alpha_{i,j}^m + \tilde{\alpha}_{i,j}^{m+1} \alpha_{i,j}^m - \tilde{\alpha}_{i,j}^{m+1} \alpha_{i,j}^m) \right\} \\
\times \exp \left\{ \sum_{m=0}^{M-1} \sum_{i,j} \beta' (\alpha_{i,j}^{m+1} - \alpha_{i,j}^m) \alpha_{i,j}^m \Delta t \right\} \\
\times \exp \left\{ \sum_{m=0}^{M-1} \left[ \alpha_{i,j}^{m+1} - \alpha_{i,j}^m \right] \right\} |n'\rangle. \tag{63} \]

Note that we have used the conventions \( \varphi_{ij}(t_m) = \varphi_{ij}^m, \alpha_{a,j}(t_m) = \alpha_{a,j}^m, \) etc. Furthermore, all ghost variables are linearly independent quantities, i.e. especially we have \( \varphi_{ij} \neq \varphi_{ji}, \psi_{ij} \neq \psi_{ji}, \omega_{ij} \neq \omega_{ji} \) and \( \tilde{\omega}_{ij} \neq \tilde{\omega}_{ji}, \) respectively.

The exchange \( i \leftrightarrow j \) in the first terms of the exponents containing the coupling between Grassmann variables and ghost variables and taking into account the symmetry \( \Theta_{ij} = \Theta_{ji} \) results in the following expression for the path integral (for \( M \to \infty \)):

\[
K(n, t; n', 0) = \tilde{N} \int \mathcal{D} \varphi \mathcal{D} \psi \mathcal{D} \omega \mathcal{D} \tilde{\omega} \exp \left\{ -\frac{1}{2} \int_0^{t'} \mathcal{D}r (\| \varphi^2 \| + \| \psi^2 \| + \| \omega^2 \| + \| \tilde{\omega}^2 \|) \right\} \\
\times \prod_{i=1}^{N} k_i(n_i, t; n_i', 0; \varphi, \psi, \omega, \tilde{\omega}), \tag{64} \]

with \( \mathcal{D} \varphi \mathcal{D} \psi \mathcal{D} \omega \mathcal{D} \tilde{\omega} = \lim_{M \to \infty} \prod_{m=1}^{M} \prod_{i,j} (\mathcal{D} \varphi_i^m \mathcal{D} \psi_j^m \mathcal{D} \omega_i^m \mathcal{D} \tilde{\omega}_j^m), \| \varphi^2 \| = \sum_{i,j} \varphi_{ij}^2(t), \| \psi^2 \| = \sum_{i,j} \psi_{ij}^2(t), \| \omega^2 \| = \sum_{i,j} \omega_{ij}^2(t), \| \tilde{\omega}^2 \| = \sum_{i,j} \tilde{\omega}_{ij}^2(t). \) The quantities \( k_i(n_i, t; n_i', 0; \)
$[\varphi, \psi, \omega, \tilde{\omega}]$ are Grassmann path integrals with respect to a single lattice site $i$:

$$k_i(n_i, t; n'_i, 0; [\varphi, \psi, \omega, \tilde{\omega}]) = \lim_{M \to \infty} \int \prod_{a=1}^{M} \prod_{m=0}^{M} (d\sigma_{a,i}^m d\sigma_{a,i}^m) \langle n_i | \exp \left\{ - (\alpha_{1,i}^M M + \alpha_{2,i}^M M) \right\}$$

$$\times \exp \left\{ \sum_{m=0}^{M-1} \sum_{j} \Theta_{ij} \left[ \sqrt{\lambda} \psi_{ji}^{m+1} - \sqrt{D} \omega_{ji}^{m+1} \right] \alpha_{1,i}^{m+1} \alpha_{1,i}^m \Delta t \right\}$$

$$\times \exp \left\{ \sum_{m=0}^{M-1} \sum_{j} \Theta_{ij} \left[ \sqrt{\beta} \psi_{ji}^{m+1} - \sqrt{\tilde{\beta}} \psi_{ij}^{m+1} \right] \alpha_{1,i}^{m+1} \alpha_{2,i}^m \Delta t \right\}$$

$$\times \exp \left\{ \sum_{m=0}^{M-1} \sum_{j} \Theta_{ij} \left[ \sqrt{\delta} \omega_{ij}^{m+1} - \sqrt{\theta} \psi_{ij}^{m+1} \right] \alpha_{2,i}^{m+1} \alpha_{2,i}^m \Delta t \right\}$$

$$\times \exp \left\{ \sum_{m=0}^{M-1} \sum_{j} \Theta_{ij} \left[ \sqrt{\lambda} \omega_{ij}^{m+1} + \sqrt{D} \omega_{ji}^{m+1} \right] \alpha_{1,i}^{m+1} \alpha_{2,i}^m \Delta t \right\}$$

$$\times \exp \left\{ \sum_{m=0}^{M-1} \sum_{j} \Theta_{ij} \left[ \sqrt{\beta} \omega_{ij}^{m+1} + \sqrt{\delta} \omega_{ji}^{m+1} \right] \alpha_{2,i}^{m+1} \alpha_{1,i}^m \Delta t \right\}$$

$$\times \exp \left\{ \sum_{m=0}^{M-1} \beta' (\alpha_{2,i}^{m+1} - \alpha_{1,i}^m) \alpha_{1,i}^m \Delta t \right\}$$

$$\times \exp \left\{ \sum_{m=0}^{M-1} \left[ \alpha_{1,i}^{m+1} - \alpha_{1,i}^m \right] \alpha_{1,i}^m + \left[ \alpha_{2,i}^{m+1} - \alpha_{2,i}^m \right] \alpha_{2,i}^m \right\} |n'_i\rangle. \quad (65)$$

Note that the kinetic interaction between the lattice sites is now transferred from the nonlinear Grassmann terms (55), (57), (59) and (61) to the terms in the exponent containing ghost variables that couple pairs of lattice sites in a linear manner. We interrupt our calculation for a short moment and focus our attention on a seemingly independent question, which becomes important for the further analysis of (65).

4. Grassmann path integrals for Ito stochastic differential equations

4.1. Ordinary linear differential equation and Fock space representation

A linear differential equation of second order can always be transformed into a system of two linear differential equations of first order. We use the representation:

$$\partial_t S = \lambda(t) Q + \beta(t) S \quad \text{and} \quad \partial_t Q = \mu(t) S + \eta(t) Q \quad (66)$$

with arbitrary time-dependent coefficients $\lambda(t)$, $\beta(t)$, $\mu(t)$ and $\eta(t)$. Note that, with respect to the following investigations, the independent variable $t$ is interpreted as time. We use the following initial conditions:

$$S(0) = S_0 \quad \text{and} \quad Q(0) = Q_0. \quad (67)$$

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It will be shown that this system can also be transformed into a representation in the reduced Fock space $\mathcal{F}_s$ with the above-introduced basis states (21). We introduce an evolution operator $\hat{E}$ associated with the system of differential equations (66):

$$\hat{E} = \lambda(t) \hat{a}_1^\dagger \hat{a}_2 + \mu(t) \hat{a}_2^\dagger \hat{a}_1 + \beta(t) \hat{a}_1^\dagger \hat{a}_1 + \eta(t) \hat{a}_2^\dagger \hat{a}_2.$$  

(68)

Using the state

$$|F(t)\rangle = S(t)|1\rangle + Q(t)|0\rangle,$$

(69)

(66) can be transformed into the evolution equation

$$\frac{\partial}{\partial t} |F(t)\rangle = \hat{E} |F(t)\rangle$$

with the initial condition

$$|F(0)\rangle = S_0|1\rangle + Q_0|0\rangle.$$  

Note that an evolution operator corresponding to the usual master equation, e.g. (29), fulfills always the identity $\langle s | \hat{F}(t) \rangle \equiv 1$, see (34) and (35), whereas this conservation becomes invalid for the more general operator (68). Here, one obtains $\langle s | \hat{F}(t) \rangle = S(t) + Q(t)$. For the following investigations, we use the notation

$$\hat{a} = \begin{pmatrix} \hat{a}_1 \\ \hat{a}_2 \end{pmatrix}, \quad \hat{a}^\dagger = (\hat{a}_1^\dagger, \hat{a}_2^\dagger), \quad \hat{A} = \begin{pmatrix} \hat{A}^+ \\ \hat{A}^- \end{pmatrix}$$

(70)

and

$$\hat{M}(t) = \begin{pmatrix} \beta(t) & \lambda(t) \\ \mu(t) & \eta(t) \end{pmatrix}, \quad \hat{E}(t) = \hat{a}^\dagger \hat{M}(t) \hat{a},$$

(71)

where $\hat{A}^+$ and $\hat{A}^-$ are defined in (39). Obviously, the solution of (66) can be represented by the formal expression

$$|F(t)\rangle = \exp \left\{ \int_0^t \hat{E}(t) \, dt \right\} |F(0)\rangle.$$  

(72)

Using the general properties (40), one obtains the averages with respect to the reduced Fock space $\mathcal{F}_s$

$$\overline{A}^+(t) = \langle s | \hat{A}^+ \exp \left\{ \int_0^t \hat{E}(t) \, dt \right\} |F(0)\rangle$$

(73)

and

$$\overline{A}^-(t) = \langle s | \hat{A}^- \exp \left\{ \int_0^t \hat{E}(t) \, dt \right\} |F(0)\rangle.$$  

(74)

These expressions may be interpreted in a twofold manner. Firstly, they are the solutions of the differential equation (66) under well-defined initial conditions contained in $|F(0)\rangle$. This can be verified from the equation of motion (38) for the averages $\overline{A}^+(t)$ and $\overline{A}^-(t)$. After some simple manipulations we arrive at

$$\frac{\partial}{\partial t} \overline{A}(t) = \langle s | \hat{A} \hat{E}(t) |F(t)\rangle = \hat{M}(t) \overline{A}(t).$$

(75)
with $\mathbf{A}(t) = [\mathbf{A}^+(t), \mathbf{A}^-(t)]$, i.e. the quantities $\mathbf{A}^+(t)$ and $\mathbf{A}^-(t)$ satisfy the same system of differential equations (66) as $S(t)$ and $Q(t)$ and they have the same initial conditions due to $\vert F(0) \rangle = S_0 \vert 1 \rangle + Q_0 \vert 0 \rangle$. The identity between $\mathbf{A}(t)$ and the solutions $[S(t), Q(t)]$ can also be checked considering the property (40). We obtain

$$\mathbf{A}^+(t) = \langle s \vert \hat{A}^+ \vert F(t) \rangle = \langle 1 \vert [S(t) \vert 1 \rangle + Q(t) \vert 0 \rangle \rangle = S(t)$$

(76)

and

$$\mathbf{A}^-(t) = \langle s \vert \hat{A}^- \vert F(t) \rangle = \langle 0 \vert [S(t) \vert 1 \rangle + Q(t) \vert 0 \rangle \rangle = Q(t).$$

(77)

Secondly, from (73), (74) and (40), it is obvious that the transition matrix

$$k(n, t; n', 0) = \langle n \vert \exp \left\{ \int_0^t \hat{E}(t) \, dt \right\} \vert n' \rangle$$

(78)

can be interpreted as the time-dependent averages $\mathbf{A}^±(t)$ with respect to the initial conditions $\mathbf{A}_0^+ = 1, \mathbf{A}_0^- = 0$ and $\mathbf{A}_0^+ = 0, \mathbf{A}_0^- = 1$, respectively, i.e.

$$\left( \begin{array}{cc} k(1, t; 1, 0) & k(1, t; 0, 0) \\ k(0, t; 1, 0) & k(0, t; 0, 0) \end{array} \right) \leftrightarrow \left( \begin{array}{cc} \mathbf{A}^+(t \vert \mathbf{A}_0^+ = 1, \mathbf{A}_0^- = 0) & \mathbf{A}^+(t \vert \mathbf{A}_0^+ = 0, \mathbf{A}_0^- = 1) \\ \mathbf{A}^-(t \vert \mathbf{A}_0^+ = 1, \mathbf{A}_0^- = 0) & \mathbf{A}^-(t \vert \mathbf{A}_0^+ = 0, \mathbf{A}_0^- = 1) \end{array} \right).$$

(79)

In other words, the transition matrix is a complete basis system of solutions with respect to the differential equations (66).

4.2. Fock space representation for Ito stochastic differential equations

In principle, the system of ordinary differential equations (66) can be generalized to Ito stochastic differential equations. Therefore, we replace the matrix $\mathbf{M}(t)$ by a deterministic term $\mathbf{M}_0(t)$ and a stochastic contribution $\sum_{\alpha=1}^{N_W} \Xi^\alpha(t) \, dW^\alpha(t) / dt$, where $dW^\alpha(t)$ are the differential components $\alpha$ of an $N_W$-dimensional Wiener process $\mathbf{W}(t)$, i.e.

$$\mathbf{M}(t) \, dt = \mathbf{M}_0 \, dt + \sum_{\alpha=1}^{N_W} \Xi^\alpha \, dW^\alpha(t).$$

(80)

Both $\mathbf{M}_0$ and $\Xi^\alpha$ may be arbitrary matrices. The Wiener process is characterized by the well-defined properties [40]:

$$dW^\alpha(t) \, dW^\beta(t) = \delta_{\alpha\beta} \, dt, \quad dW^\alpha(t) \, dt = 0 \quad \text{and} \quad [dW^\alpha(t)]^{N+2} = 0 \quad (N > 0).$$

(81)

With $\mathbf{M}_0(t)$ from (80), the equation $\delta \vert F(t) \rangle = \hat{E}(t) \vert F(t) \rangle$ now becomes an Ito stochastic differential equation

$$d \vert F(t) \rangle = \hat{E}(t) \, dt \vert F(t) \rangle = \left[ \hat{a}^\dagger \mathbf{M}_0 \hat{a} \, dt + \sum_{\alpha=1}^{N_W} (\hat{a}^\dagger \Xi^\alpha \hat{a}) \, dW^\alpha(t) \right] \vert F(t) \rangle$$

(82)
for the Fock state $|F(t)\rangle$ and the corresponding evolution of the averages (75) must be replaced by

$$d\overline{A}(t) = M_0 A(t) \, dt + \sum_{\alpha=1}^{N_W} (\Xi^\alpha \overline{A}(t) ) \, dW_\alpha(t),$$

which is the corresponding Ito stochastic differential equation for the time-dependent average $\overline{A}(t)$. For the further analysis, it is an important remark at this point that the introduced averages are only performed with respect to the states of the reduced Fock space, while the Wiener processes are not concerned by this averaging procedure. Furthermore, the state $|F(t)\rangle$ can be explicitly written as

$$|F(t)\rangle = A^+(t)|1\rangle + A^-(t)|0\rangle$$

as follows from the comparison with (69), (76) and (77).

4.3. Grassmann path integrals for Ito stochastic differential equations

4.3.1. Infinitesimal time step: Fermionic algebra. The aim of the following investigation is the representation of the solution of an Ito stochastic differential equation in terms of a Grassmann path integral. The first step is the determination of a time-dependent operator $\hat{E}'(t)$ describing the infinitesimal evolution:

$$|F(t) + dt\rangle = |F(t)\rangle + d|F(t)\rangle = \exp(\hat{E}'(t) \, dt)|F(t)\rangle.$$  

The operator $\hat{E}'(t)$ can be obtained from a comparison of (82) and (85). Hence we get

$$\exp(\hat{E}'(t) \, dt) = 1 + \hat{E}(t) \, dt = 1 + \hat{a}^\dagger M_0 \hat{a} \, dt + \sum_{\alpha=1}^{N_W} (\hat{a}^\dagger \Xi^\alpha \hat{a} ) \, dW_\alpha(t) + \gamma \hat{a}^\dagger \hat{a}_1 \hat{a}_2^\dagger \, dt.$$  

The introduction of the additional term $\gamma \hat{a}^\dagger \hat{a}_1 \hat{a}_2^\dagger \, dt$ is always possible because of the identity (41). After some algebraic transformations (see appendix A), we arrive at the following expression for $\hat{E}'(t)$:

$$\hat{E}'(t) \, dt = \hat{a}^\dagger \left[ M_0 - \frac{1}{2} \sum_\alpha (\Xi^\alpha \Xi^\dagger ) \right] \hat{a} \, dt + \sum_\alpha (\hat{a}^\dagger \Xi^\alpha \hat{a} ) \, dW_\alpha(t)$$

$$+ \left( \gamma - \sum_\alpha \det |\Xi^\alpha| \right) \hat{a}^\dagger \hat{a}_1 \hat{a}_2^\dagger \, dt.$$  

The difference between $\hat{E}'(t)$ and $\hat{E}(t)$ stems from the fact that a Wiener process scales as $dW_\alpha(t) \sim \sqrt{dt}$.

4.3.2. Infinitesimal time step: Grassmann algebra. The next step is the transformation of the infinitesimal evolution (85) into a Grassmann path integral. For that purpose, we replace the creation and annihilation operators by Grassmann variables and the corresponding differentiation
operators, see (49). Therefore, (87) becomes
\[
\dot{A}(t) \, dt = \sum_{a,b} \bar{a}_a \left[ (M_{ab}^0 - \frac{1}{2} \sum_{a,c} \Xi_{ac}^{\alpha} \Xi_{cb}^{\alpha}) \, dt + \sum_{a} \Xi_{ab}^{\alpha} \, dW_a(t) \right] \frac{\partial}{\partial \bar{a}_b} + \left( \gamma - \sum_{a} \text{det}[\Xi^{\alpha}] \right) \, dt \bar{a}_1 \frac{\partial}{\partial \bar{a}_1} \bar{a}_2 .
\] (88)

Furthermore, the representation (84) of the evolution state \(|F(t)\rangle\) is given in the explicit form
\[
|F(t)\rangle = \widetilde{A}^+(t)\bar{a}_1 + \widetilde{A}^-(t)\bar{a}_2 .
\] (89)

The substitution of (88) into (85) leads to the Grassmann representation of the solution of an Ito stochastic differential equation (see appendix B for details). One obtains
\[
\langle F(t + dt), \bar{a} \rangle = \int \prod_{a=1}^{2} (d\bar{a}_a \, d\dot{a}_a) \exp \left\{ E''(t, \bar{a}, \dot{a}') \, dt - \sum_{a=1}^{2} \dot{a}_a' (\bar{a}_a - \bar{a}_a') \right\} \langle F(t), \bar{a} \rangle .
\] (90)

Note that we have here used the representation \(|F(dt), \bar{a}\rangle\) instead of \(|F(dr)\rangle\) in order to show explicitly the dependence on the Grassmann variables. The function \(E''\) is given by
\[
E''(t, \bar{a}, \dot{a}') \, dt = \sum_{a,b} \bar{a}_a \left[ M_{ab}^0 \, dt + \sum_{a} \Xi_{ab}^{\alpha} \, dW_a(t) \right] \dot{a}_b' .
\] (91)

Note that the matrices \(M_0^0\) and \(\Xi^{\alpha}\) are identical to those in (83). It should be remarked that \(E''(t, \bar{a}, \dot{a}')\) is not a completely unique expression. Without changing the physical content, this quantity may be modified by the addition of an arbitrary term proportional to \(\bar{a}_1 \dot{a}_1' \bar{a}_2 \dot{a}_2'\). This possible gauge is a result of the original representation in the reduced Fock space \(\mathcal{F}_s\), see (41).

### 4.3.3. Grassmann path integral.

The successive continuation of the above-discussed infinitesimal steps leads straightforwardly to the following result:
\[
\langle F(t_M), \{ \bar{a}^M \} \rangle = \int \prod_{m=0}^{M-1} \prod_{a=1}^{2} (d\bar{a}_a^m \, d\dot{a}_a^m) \exp \left\{ \sum_{m=0}^{M-1} \left[ E''(t_{m+1}, \bar{a}^{m+1}, \dot{a}^m) \, dt - \dot{a}^m (\bar{a}^{m+1} - \bar{a}^m) \right] \right\} \times \langle F(0), \{ \bar{a}^0 \} \rangle .
\] (92)

The discrete timescale is chosen as \(t_m = m \, dt\) and we have used again the convention \(\bar{a}^m = \bar{a}(t_m)\). The time dependence of \(E''(t)\) stems from the stochastic Wiener processes. The discrete version of a Wiener process can be written as \(dW(t) \rightarrow \Delta W(t_{m+1}) = W(t_{m+1}) - W(t_m)\), thus \(E''(t)\) in (92) must be replaced by \(E''(t_{m+1})\).

\[1\] Obviously, the set of zero operators in \(\mathcal{F}_s\) (i.e. operators with the property \(\hat{C}|\bar{w}\rangle = 0\) for any state \(|\bar{w}\rangle \in \mathcal{F}_s\)) contains more elements than the set of zero operators related to the complete Fock space \(\mathcal{F}\) (e.g. \(\hat{A}^+_i \hat{A}^-_j |\bar{w}\rangle = 0\) for \(|\bar{w}\rangle \in \mathcal{F}\), but there exists at least one state \(|\bar{w}'\rangle \in \mathcal{F}\) with \(\hat{A}^+_i \hat{A}^-_j |\bar{w}'\rangle \neq 0\)). In principle, the complete Fock space has only the trivial zero operator \(\hat{0}\), with the exception of such combinations of operators that can be identically transformed into \(\hat{0}\) by using the Fermionic anticommutation rules.
Now we are able to determine the transition matrix (78) as a Grassmann path integral. Considering that the scalar product using Grassmann variables is defined by
\[
\langle b | b' \rangle = \int \prod_{a=1}^{2} (\text{d} \alpha_a \text{d} \alpha'_a) \langle b, \{ \alpha \} | \exp \{ \alpha \bar{\alpha} \} | b', \{ \bar{\alpha} \} \rangle,
\]
(93)
one obtains immediately
\[
k(n; t; n', 0) = \int \prod_{m=0}^{M} \prod_{a=1}^{2} (\text{d} \alpha_a^m \text{d} \alpha'_a^m) \langle n, \{ \alpha \} | \exp \{ \alpha^M \bar{\alpha}^M \} \rangle \times \exp \left\{ \sum_{m=0}^{M-1} \left[ E''(t_{m+1}, \bar{\alpha}^{m+1}, \alpha^m) \text{d} t - \alpha^m (\bar{\alpha}^{m+1} - \bar{\alpha}^m) \right] \right\} \times |n', \{ \bar{\alpha}^0 \} \rangle.
\]
(94)
We have now arrived at an important point of our discussion. Let us assume that we have a path integral of type (96). Then, due to (79), we can identify the elements of the transition matrix \(k(n; t; n', 0)\) as solutions \(\overline{A}(t)\) of the system of stochastic differential equations (83) with well-defined initial conditions, namely
\[
\overline{A}(0) = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad \text{for } n' = 1 \quad \text{and} \quad \overline{A}(0) = \begin{pmatrix} 0 \\ 1 \end{pmatrix} \quad \text{for } n' = 0.
\]
(95)
Therefore, we obtain the following alternative representation of the transition matrix (79)
\[
k(n; t; n', 0; W) = \langle n | \exp \left\{ \int_0^t \hat{E}(t) \text{d} t \right\} | n' \rangle = \overline{A}^+(t|n') \overline{A}^-(t|n')^{-1-n},
\]
(96)
where \(\overline{A}(t|n')\) indicates that this solution belongs to the initial condition corresponding to the map (95). We remind that the quantities \(\overline{A}^\pm(t)\) are still stochastic observables depending on the actual realization of the underlying Wiener processes. Thus the transition matrix \(k(n; t; n', 0; W)\) depends also on this special Wiener process \(W\). The notation \(\overline{A}\) indicates only that these quantities are averages with respect to the states of the original reduced Fock space \(\mathcal{F}_s\).

4.4. Averages with respect to Wiener processes

Usually, the solution of an Ito stochastic differential equation for a special realization \(W(t)\) of the Wiener process is not of interest. However, averages over all realizations play an important role. Therefore, we give here the formal average of the transition matrix with respect to the Wiener processes
\[
\langle k(n; t; n', 0; [W]) \rangle_w = \langle n | \exp \left\{ \int_0^t \hat{E}(t) \text{d} t \right\} | n' \rangle_w,
\]
(97)
which we need for our further discussions. Using (96), the averaged matrix elements of (97) correspond to the mean quantities \(\langle \overline{A}^\pm(t) \rangle\) with respect to the multi-dimensional Wiener process \(W\) (with dimension \(N_w\)). The introduction of normalized noise terms \(\chi_a\) with \(\text{d} W_a(t) = \chi_a(t) \text{d} t\) leads to
\[
\langle \chi_a(t) \chi_b(t) \text{d} t^2 \rangle = \langle \text{d} W_a(t) \text{d} W_b(t) \rangle = \delta_{ab} \text{d} t.
\]
(98)
Thus the multi-dimensional Wiener process $W$ with the components $W_\alpha$ ($\alpha = 1, \ldots, N_W$) corresponds to a normalized Gaussian measure with the probability distribution

$$P_G [x] = \lim_{M \to \infty} \left( \frac{dt}{2\pi} \right)^{N_W/2} \exp \left\{ -\frac{1}{2} \sum_{m=1}^{M} \sum_{\alpha=1}^{N_W} x_\alpha(t_m)^2 dt \right\}$$

$$= \mathcal{N} \lim_{M \to \infty} \exp \left\{ -\frac{1}{2} \sum_{m=1}^{M} |x(t_m)|^2 dt \right\}$$

(99)

with the normalization prefactor $\mathcal{N}$.

5. Stochastic differential equations for reaction–diffusion systems

5.1. Stochastic evolution equation

Now we come back to the original problem and continue the discussion of the path integral (65). Obviously, this path integral can be interpreted as a product

$$\prod_{i=1}^{N} k_i(n_i, t; n'_i, 0; [\varphi, \psi, \omega, \tilde{\omega}])$$

averaged over a set of normalized Wiener processes (given by the quantities $\varphi_{ij}$, $\psi_{ij}$, $\omega_{ij}$ and $\tilde{\omega}_{ij}$ or, more explicitly, $dW_{\varphi}^{ij}(t) = \varphi_{ij}(t) dt$, $dW_{\psi}^{ij}(t) = \psi_{ij}(t) dt$, $dW_{\omega}^{ij}(t) = \omega_{ij}(t) dt$ and $dW_{\tilde{\omega}}^{ij}(t) = \tilde{\omega}_{ij}(t) dt$, respectively). Thus (65) can be rewritten as

$$K(n, t; n', 0) = \left( \prod_{i=1}^{N} k_i(n_i, t; n'_i, 0; [\varphi, \psi, \omega, \tilde{\omega}]) \right)_{\varphi, \psi, \omega, \tilde{\omega}} .$$

(100)

The quantities $k_i(n_i, t; n'_i, 0; [\varphi, \psi, \omega, \tilde{\omega}])$ are path integrals depending on the actual Wiener processes. The comparison between (65) and the general mathematical structure of (91) shows that $k_i$ can be represented by (96), i.e.

$$k_i(n_i, t; n'_i, 0; [\varphi, \psi, \omega, \tilde{\omega}]) = A_{i}^+(t | n'_i) n_i A_{i}^-(t | n'_i) ,$$

(101)

where $A_{i}^+(t | n'_i)$ and $A_{i}^-(t | n'_i)$ are solutions of the corresponding system of Ito stochastic differential equations:

$$dA_{i}^+(t) = M_{i,0} A_{i}^+(t) dt + \sum_{m,n} \left( \Xi_{i,mn}^{\varphi} A_{i}(t) \right) dW_{mn}^\varphi(t) + \sum_{m,n} \left( \Xi_{i,mn}^{\psi} A_{i}(t) \right) dW_{mn}^\psi(t)$$

$$+ \sum_{m,n} \left( \Xi_{i,mn}^{\omega} A_{i}(t) \right) dW_{mn}^\omega(t) + \sum_{m,n} \left( \Xi_{i,mn}^{\tilde{\omega}} A_{i}(t) \right) dW_{mn}^{\tilde{\omega}}(t)$$

(102)

with the initial conditions (95). The comparison of (65), (91) and (94) leads to the matrices:

$$M_{i,0} = \beta' \begin{pmatrix} -1 & 0 \\ 1 & 0 \end{pmatrix}$$

(103)

$$\Xi_{i,mn}^{\varphi} = \Theta_{mn} \sqrt{\lambda} \begin{pmatrix} \delta_{im} & \delta_{im} \\ 0 & -\delta_{im} \end{pmatrix} , \quad \Xi_{i,mn}^{\psi} = \Theta_{mn} \sqrt{\beta} \begin{pmatrix} \delta_{im} - \delta_{im} & 0 \\ \delta_{im} & 0 \end{pmatrix}$$

(104)

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controlling the strength of the coupling between the Wiener processes and the dynamic variables $\Xi_i(t)$. The structure of these matrices is mainly determined by the underlying elementary processes describing diffusion, creation and annihilation processes. It seems to be simple to construct the corresponding matrices also for other elementary unimolecular and bimolecular reactions and diffusion processes because the general concept is the same.

5.2. Averages and correlation functions

The knowledge of the transition matrix $K(n, t; n', 0)$ (see equation 100) allows the determination of arbitrary averages, moments and correlation functions. In the following investigations, we confine ourselves to averages and correlation functions of the local occupation numbers $\hat{A}^+_i$, see (39). All other quantities can be determined by an analogous procedure. Using (37), we obtain

$$\langle A^+_i(t) \rangle = \langle s| \hat{A}^+_i|F(t)\rangle = \sum_m \langle m| \hat{A}^+_i|F(t)\rangle. \quad (106)$$

Because of the identity $\langle m| \hat{A}^+_i \rangle = \langle m| \delta_{mi, 1} \rangle$, we have with (44) and (45)

$$\langle A^+_i(t) \rangle = \sum_m \delta_{m, 1} \langle m|F(t)\rangle = \sum_{m, n'} \delta_{m, 1} K(n, t; n', 0) P(n', 0) \quad (107)$$

or with (100) and (101)

$$\langle A^+_i(t) \rangle = \sum_{m, n'} \delta_{m, 1} \left( \prod_{j=1}^N \left[ \hat{A}^+_j(t|n'_j)^{m_j} \hat{A}^-_j(t|n'_j)^{1-m_j} \right] \right)_{\psi, \psi', \omega, \tilde{\omega}} P(n', 0), \quad (108)$$

where the quantities $\hat{A}^+_j(t|n'_j)$ and $\hat{A}^-_j(t|n'_j)$ are the above-introduced solutions of the stochastic differential equations (102) subject to the special initial conditions (95). An important aspect becomes clear in the last equation. The left-hand side $\langle A^+_i(t) \rangle$ is the local occupation density of the lattice site $i$ for the diffusion–reaction system with exclusion restrictions. Obviously, this quantity contains implicitly the interaction with neighboured lattice sites due to the evolutionary rules. The right-hand side of (109) is a product of quantities, which are apparently independent solutions of stochastic differential equations (102) for each lattice site. The coupling between these quantities is organized by the averaging procedure with respect to the Wiener processes $W^\psi_{ij}(t)$, $W^\psi_{ji}(t)$, $W^\omega_{ij}(t)$ and $W^\tilde{\omega}_{ij}(t)$, because the stochastic processes which drive the evolution of the Ito differential equations of neighboured lattice points are not completely independent. Hence, the average procedure connects now the lattice sites of the system. The summation over all configurations can be carried out and we get

$$\langle A^+_i(t) \rangle = \sum_{n'} \left( \prod_{j \neq i}^{N} \left[ \hat{A}^+_j(t|n'_j)^{N} \left( \hat{A}^+_j(t|n'_j)^{N} + \hat{A}^-_j(t|n'_j) \right) \right] \right)_{\psi, \psi', \omega, \tilde{\omega}} P(n', 0). \quad (109)$$
A similar calculation leads to higher moments, e.g.

\[
\langle A_i^+(t) A_j^+(t) \rangle = \sum_{n'} \left( A_i^+(t|n') A_j^+(t|n') \prod_{k \neq i,j} [A_k^+(t|n') + A_k^-(t|n')] \right) P(n', 0).
\]  \quad (110)

Correlation functions can be represented by similar expressions, e.g.

\[
\langle A_i^+(t) A_j^+(0) \rangle = \sum_{n'} \left( A_i^+(t|n') \prod_{k \neq i} [A_k^+(t|n') + A_k^-(t|n')] \right) \delta_{n',1} P(n', 0).
\]  \quad (111)

Considering that the initial conditions may also be written as

\[
A_k^+(0|n'_k) = \delta_{n'_k,1} \quad \text{and} \quad A_k^-(0|n'_k) = \delta_{n'_k,0},
\]  \quad (112)

(111) can be identically transformed into

\[
\langle A_i^+(t) A_j^+(0) \rangle = \sum_{n'} \left( A_i^+(t|n') \prod_{k \neq i} [A_k^+(t|n') + A_k^-(t|n')] \right) P(n', 0).
\]  \quad (113)

Note that the initial conditions require \( A_k^+(0|n'_k) + A_k^-(0|n'_k) = 1 \), whereas the solution of the system of stochastic differential equations (104) is usually characterized by the property \( A_k^+(t|n'_k) + A_k^-(t|n'_k) \neq 1 \) for \( t > 0 \).

6. Fokker–Planck equation

6.1. Ito stochastic differential equations and Fokker–Planck equation

The determination of averages and correlation functions using (107), (110) and (113) and the solution of the corresponding stochastic differential equations (e.g. by Monte Carlo simulations) is very hard, because of the complicated mathematical structure of (107), (110) and (113). The difficulties stem, on the one hand, from the Wiener processes acting simultaneously at pairs of lattice points. On the other, the local averages \( \langle A_i^+(t) \rangle \) and the two site correlation functions \( \langle A_i^+(t) A_j^+(0) \rangle \) depend on the time evolution of \( A_k^+(t) \) and \( A_k^-(t) \) at all lattice sites, see (107), (110) and (113).

There exists always a probability \( P(\overline{A}, t; \overline{A}_0, 0) \) determining the transition from the initial values \( \overline{A}_0 \) (at time \( t = 0 \)) to the final values \( \overline{A} \) (at time \( t \)) under the influence of multi-dimensional Wiener processes \( W^\varphi, W^\psi, W^\omega \) and \( W^\tilde{\omega} \) via the Ito stochastic differential equation (102). The generating equation for this probability can be obtained from the well-known connection between Ito stochastic differential equations and Fokker–Planck equations [40]. One obtains the following
A simple algebraic calculation considering (107) leads to the representation:

$$\frac{\partial}{\partial t} P(\mathbf{A}, t; \mathbf{A}_0, 0) = \frac{1}{2} \sum_{ij,abcd} \left\{ \frac{\partial}{\partial A_{a,i}} \frac{\partial}{\partial A_{b,j}} \sum_{mn} \left[ \Xi_{i,mn}^{acw} \Xi_{j,mn}^{bdo} + \Xi_{i,mn}^{acw} \Xi_{j,mn}^{bdo} \right] A_{c,i} A_{d,j} P(\mathbf{A}, t; \mathbf{A}_0, 0) \right\} $$

$$+ \frac{1}{2} \sum_{ij,abcd} \left\{ \frac{\partial}{\partial A_{a,i}} \frac{\partial}{\partial A_{b,j}} \sum_{mn} \left[ \Xi_{i,mn}^{acw} \Xi_{j,mn}^{bdo} + \Xi_{i,mn}^{acw} \Xi_{j,mn}^{bdo} \right] A_{c,i} A_{d,j} P(\mathbf{A}, t; \mathbf{A}_0, 0) \right\} $$

$$+ \sum_{i,ab} \left\{ \frac{\partial}{\partial A_{a,i}} M_{i,0} A_{b,j} P(\mathbf{A}, t; \mathbf{A}_0, 0) \right\}. \tag{114}$$

$\Xi_{i,mn}^{acw}, \Xi_{i,mn}^{acw}, \Xi_{j,mn}^{bdo}$ and $\Xi_{j,mn}^{bdo}$ are the matrix elements of $\Xi_{i,mn}^{acw}, \Xi_{i,mn}^{acw}, \Xi_{j,mn}^{bdo}$ and $\Xi_{j,mn}^{bdo}$, respectively. A simple algebraic calculation considering (107) leads to the representation:

$$\frac{\partial}{\partial t} P(\mathbf{A}, t; \mathbf{A}_0, 0) = -\beta' \sum_i \left( \frac{\partial}{\partial A_i} - \frac{\partial}{\partial A_i} \right) A_i P(\mathbf{A}, t; \mathbf{A}_0, 0) $$

$$+ \frac{z\lambda}{2} \sum_i \left[ \left( \frac{\partial}{\partial A_i} \right)^2 (\mathbf{A}_i)^2 + \left( \frac{\partial}{\partial A_i} - \frac{\partial}{\partial A_i} \right)^2 (\mathbf{A}_i)^2 \right] P(\mathbf{A}, t; \mathbf{A}_0, 0) $$

$$+ \lambda \sum_{ij} \Theta_{ij} \frac{\partial}{\partial A_i} \left( \frac{\partial}{\partial A_j} - \frac{\partial}{\partial A_j} \right) A_i A_j P(\mathbf{A}, t; \mathbf{A}_0, 0) $$

$$+ \frac{z\beta}{2} \sum_i \left[ 2 \frac{\partial}{\partial A_i} \left( \frac{\partial}{\partial A_i} - \frac{\partial}{\partial A_i} \right) + \left( \frac{\partial}{\partial A_i} \right)^2 \right] (\mathbf{A}_i)^2 P(\mathbf{A}, t; \mathbf{A}_0, 0) $$

$$+ \beta \sum_{ij} \Theta_{ij} \frac{\partial}{\partial A_i} \left[ \frac{\partial}{\partial A_j} - \frac{\partial}{\partial A_j} \right] A_i A_j P(\mathbf{A}, t; \mathbf{A}_0, 0) $$

$$+ \frac{zD}{2} \sum_i \left\{ \left[ \left( \frac{\partial}{\partial A_i} \right)^2 \right]^2 + \left( \frac{\partial}{\partial A_i} \right)^2 \right\} (\mathbf{A}_i)^2 P(\mathbf{A}, t; \mathbf{A}_0, 0) $$

$$+ D \sum_{ij} \Theta_{ij} \frac{\partial}{\partial A_i} \frac{\partial}{\partial A_j} [\mathbf{A}_i A_j - \mathbf{A}_j A_i] P(\mathbf{A}, t; \mathbf{A}_0, 0), \tag{115}$$

where $z$ is the coordination number of the underlying lattice.

6.2. Probability and pseudo-probability

6.2.1. Pseudo-probability. Equation (115) allows the determination of the probability distribution $P(\mathbf{A}, t; \mathbf{A}_0, 0)$. Thus, averages (107) and (110) and correlation functions (113) can be obtained directly from this probability. In terms of this probability, the relevant averages are now defined by

$$\left\langle A_i^j (t|n_i') \prod_{j \neq i} A_j^j (t|n_j') \right\rangle_{\Phi, \omega, \tilde{\omega}} $$

$$= \int \prod_{k=1}^N (d\mathbf{A}_k' d\mathbf{A}_k') A_i^k \prod_{j \neq i} A_j^j \left[ A_i^j + A_j^i \right] P(\mathbf{A}, t; \mathbf{A}_0, 0). \tag{116}$$
A real disadvantage is the remaining product on the right-hand side, because it requires the
calculation of high moments in order to obtain the simple local occupation rate \( \langle A_i^+ (t) \rangle \) of the
underlying diffusion reaction lattice system, see also (109). A similar situation occurs also for
the determination of other moments or correlation functions. All these quantities contain similar
product terms, see (109), (110) and (113). Thus, it seems to be reasonable to introduce

\[
G(\vec{A}, t; \vec{A}_0, 0) = \prod_j^{N} [\overline{A}_j^+ + \overline{A}_j^-] P(\vec{A}, t; \vec{A}_0, 0),
\]

(117)

which leads to the more favourable representation:

\[
\left\langle A_i^+ (t_n) \prod_{j \neq i}^{N} [\overline{A}_j^+ (t_{n'}) + \overline{A}_j^- (t_{n''})] \right\rangle \approx \int \prod_{k=1}^{N} (d\overline{A}_k^+ d\overline{A}_k^-) \frac{\overline{A}_i^-}{\overline{A}_i^+ + \overline{A}_i^-} G(\vec{A}, t; \vec{A}_0, 0). \quad (118)
\]

The new quantity \( G(\vec{A}, t; \vec{A}_0, 0) \) is no longer a real probability distribution function, although
it plays the role of a probability in the formulae for moments and correlation functions, see
for example (118). Therefore, let us speak about a pseudo-probability (117). An important
cause why \( G(\vec{A}, t; \vec{A}_0, 0) \) is not a probability follows from the fact that this quantity is not
positive definite, see (117). However, this pseudo-probability can be generated also by a partial
differential equation similar to a Fokker–Planck equation. The insertion of \( P(\vec{A}, t; \vec{A}_0, 0) =
G(\vec{A}, t; \vec{A}_0, 0)/\prod_j^{N} [\overline{A}_j^+ + \overline{A}_j^-] \) into (115) and simple algebraic transformations lead to

\[
\frac{\partial}{\partial t} G = \sum_i \left[ \beta' \left( \frac{\partial}{\partial \overline{A}_i^-} - \frac{\partial}{\partial \overline{A}_i^+} \right) \overline{A}_i^+ - z \left( \lambda + \beta \right) \frac{\partial}{\partial \overline{A}_i^+} \left( \overline{A}_i^+ \right)^2 \right] G
\]

\[-z D \sum_i \left[ \left( \frac{\partial}{\partial \overline{A}_i^+} - \frac{\partial}{\partial \overline{A}_i^-} \right) \left( \overline{A}_i^+ \right)^2 + \left( \frac{\partial}{\partial \overline{A}_i^+} - \frac{\partial}{\partial \overline{A}_i^-} \right) \left( \overline{A}_i^- \right)^2 \right] G \]

\[+ \sum_{ij} \Theta_{ij} \left( \frac{\partial}{\partial \overline{A}_i^-} - \frac{\partial}{\partial \overline{A}_i^+} \right) \beta \overline{A}_j^+ \overline{A}_i^- - \lambda \overline{A}_j^- \overline{A}_i^+ - D \left( \overline{A}_i^- \overline{A}_j^- - \overline{A}_i^+ \overline{A}_j^+ \right) G \]

\[+ \frac{z \lambda}{2} \sum_i \left[ \left( \frac{\partial}{\partial \overline{A}_i^+} \right)^2 + \left( \frac{\partial}{\partial \overline{A}_i^-} \right)^2 \right] \left( \overline{A}_i^+ \right)^2 G \]

\[+ \frac{z \beta}{2} \sum_i \left[ \left( \frac{\partial}{\partial \overline{A}_i^-} \right)^2 + \left( \frac{\partial}{\partial \overline{A}_i^-} \right)^2 \right] \left( \overline{A}_i^- \right)^2 G \]

\[+ \frac{z D}{2} \sum_i \left[ \left( \frac{\partial}{\partial \overline{A}_i^+} \right)^2 + \left( \frac{\partial}{\partial \overline{A}_i^-} \right)^2 \right] \left( \overline{A}_i^+ \right)^2 G \]

\[+ \sum_{ij} \Theta_{ij} \frac{\partial}{\partial \overline{A}_i^-} \left[ \left( \frac{\partial}{\partial \overline{A}_j^+} - \frac{\partial}{\partial \overline{A}_j^-} \right) \beta \overline{A}_j^+ - \lambda \overline{A}_j^- \right] + D \frac{\partial}{\partial \overline{A}_j^-} \left[ \overline{A}_i^- \overline{A}_j^- - \overline{A}_i^+ \overline{A}_j^+ \right] \right] G \quad (119)
\]

with the functional dependence \( G = G(\vec{A}, t; \vec{A}_0, 0) \). This linear partial differential equation has
a structure similar to the usual Fokker–Planck equation.
6.2.2. Transformation of variables. The pseudo-probability \( G(\overline{A}, t; \overline{A}_0, 0) \) simplifies considerably the determination of averages and correlation functions, but the averaging procedure must be performed now over a fraction, see (118). Therefore, a mapping \( \{ \overline{A}_i^+, \overline{A}_i^- \} \rightarrow \{ N_i, \phi_i \} \) defined by

\[
\overline{A}_i^+ = N_i \phi_i \quad \text{and} \quad \overline{A}_i^- = N_i (1 - \phi_i)
\]  

seems to be reasonable. This transformation leads to a new representation of (119) for the pseudo-probability \( G(\overline{A}, t; \overline{A}_0, 0) \rightarrow G(N, \phi, t; N_0, \phi_0, 0) \). One obtains

\[
\frac{\partial}{\partial t} G = \sum_i \frac{\partial}{\partial \phi_i} \left\{ \beta' \phi_i + z (\lambda + \beta) \phi_i^2 (1 - \phi_i) + z D (1 - 2 \phi_i) \left[ \phi_i^2 + (1 - \phi_i)^2 \right] \right\} G
\]

\[
+ \frac{z \lambda}{2} \sum_i \left\{ \frac{\partial^2}{\partial N_i^2} \phi_i^2 + 2 \frac{\partial^2}{\partial N_i \partial \phi_i} N_i \phi_i^2 (1 - \phi_i) + \frac{\partial^2}{\partial \phi_i^2} (1 - \phi_i)^2 (1 + \phi_i) \right\} G
\]

\[
+ \frac{z \beta}{2} \sum_i \left\{ \frac{\partial^2}{\partial N_i^2} \phi_i^2 + 2 \frac{\partial^2}{\partial N_i \partial \phi_i} N_i \phi_i^2 (1 - \phi_i) + \frac{\partial^2}{\partial \phi_i^2} \phi_i^2 (1 + (1 - \phi_i)^2) \right\} G
\]

\[
+ \sum_{ij} \Theta_{ij} \left\{ \left[ \frac{\partial}{\partial N_j} N_i \phi_i + \frac{\partial}{\partial \phi_i} \phi_j (1 - \phi_i) \right] (\lambda (1 - \phi_j) - \beta \phi_j) \right\} G
\]

\[
+ \sum_{ij} \Theta_{ij} \left[ \frac{\partial^2}{\partial N_i^2} \phi_i + \frac{\partial^2}{\partial N_j \partial \phi_i} \phi_j (1 - \phi_i) \right] \left[ \phi_i^2 + (1 - \phi_i)^2 \right] G
\]

\[
+ \sum_{ij} \Theta_{ij} \left[ \frac{\partial}{\partial N_j} N_i N_j + \frac{\partial}{\partial N_j} \frac{\partial}{\partial \phi_i} N_j - \frac{\partial}{\partial \phi_i} \frac{\partial}{\partial \phi_j} (1 - \phi_i) \phi_j \right] (\phi_j - \phi_i) G.
\]  

From (120), the initial conditions are now defined by \( N_{i,0} \equiv 1 \) and \( \phi_{i,0} = n'_i \).

6.2.3. New probability distribution. The transformation of the variables (120) changes also the integral measure on the right-hand side of (118). Because of

\[
\prod_{k=1}^{N} (d \overline{A}_k^+ d \overline{A}_k^-) = \prod_{k=1}^{N} \left( \begin{array}{c} N_k \\ \phi_k \\ 1 - \phi_k \end{array} \right) \left( \begin{array}{c} N_k \\ d N_k \end{array} \right) = \prod_{k=1}^{N} (N_k d N_k d \phi_k),
\]  

one obtains instead of (118)

\[
\left\langle \overline{A}_i^+ (t, \phi_i) \prod_{j \neq i}^{N} \left[ \overline{A}_j^+ (t, \phi_j) + \overline{A}_j^- (t, \phi_j) \right] \right\rangle_{\phi_0 \phi} = \int \prod_{k=1}^{N} (d N_k d \phi_k) \phi_j \left( \prod_{k=1}^{N} N_k \right) G(N, \phi, t; N_0, \phi_0, 0).
\]  

\[\text{(123)}\]
Equation (118) suggests the introduction of a new distribution function:

$$\Psi(N,\phi,t;N_0,\phi_0,0) = \left(\prod_{k=1}^{N} N_k\right) G(N,\phi,t;N_0,\phi_0,0). \quad (124)$$

In contrast to $G(N,\phi,t;N_0,\phi_0,0)$, the function $\Psi(N,\phi,t;N_0,\phi_0,0)$ is positive definite ($\Psi = (\prod_{k=1}^{N} N_k)^2 P$, see also (117)). Furthermore, the function $\Psi$ is a solution of the following partial differential equation:

$$\frac{\partial}{\partial t} \Psi = \beta' \sum_{i} \frac{\partial}{\partial \phi_i} \phi_i \Psi$$

$$+ \frac{z\lambda}{2} \sum_{i} \left[ \frac{\partial^2 N_i^2 \phi_i^2 \Psi}{\partial N_i^2} - 2 \frac{\partial N_i \phi_i^2 \Psi}{\partial N_i} + 2 \frac{\partial^2 N_i \phi_i^2 (1 - \phi_i) \Psi}{\partial N_i \partial \phi_i} + \frac{\partial^2 (1 - \phi_i)^2 (1 + \phi_i^2) \Psi}{\partial \phi_i^2} \right]$$

$$+ \frac{z\beta}{2} \sum_{i} \left[ \frac{\partial^2 N_i^2 \phi_i^2 \Psi}{\partial N_i^2} - 2 \frac{\partial N_i \phi_i^2 \Psi}{\partial N_i} + 2 \frac{\partial^2 N_i \phi_i^2 (1 - \phi_i) \Psi}{\partial N_i \partial \phi_i} + \frac{\partial^2 (2 \phi_i^2 - 2 \phi_i^2 + \phi_i^4) \Psi}{\partial \phi_i^2} \right]$$

$$+ \tilde{\lambda} \sum_{ij} \Theta_{ij} \left[ \frac{\partial^2 N_i \phi_i (1 - \phi_j) \Psi}{\partial N_i \partial \phi_j} - \frac{\partial \phi_i (1 - \phi_j) \Psi}{\partial \phi_j} + \frac{\partial^2 (1 - \phi_i) \phi_j (1 - \phi_j) \Psi}{\partial \phi_i \partial \phi_j} \right]$$

$$- \beta \sum_{ij} \Theta_{ij} \left[ \frac{\partial^2 N_i \phi_i \phi_j \Psi}{\partial N_i \partial \phi_j} - \frac{\partial \phi_i \phi_j \Psi}{\partial \phi_j} + \frac{\partial^2 (1 - \phi_i) \phi_j \phi_j \Psi}{\partial \phi_i \partial \phi_j} \right]$$

$$+ zD \sum_{i} \left\{ \frac{\partial^2 N_i^2 \phi_i^2 (1 - \phi_i)^2 \Psi}{\partial N_i^2} - 2 \phi_i (1 - \phi_i)^2 \Psi \right\}$$

$$+ \frac{zD}{2} \sum_{i} \left\{ 2 \frac{\partial^2 N_i (1 - 2 \phi_i) \phi_i^2 + (1 - \phi_i)^2 \Psi}{\partial N_i \partial \phi_i} + \frac{\partial^2 \phi_i^2 + (1 - \phi_i)^2 \Psi}{\partial \phi_i^2} \right\}$$

$$+ D \sum_{ij} \Theta_{ij} \left\{ \frac{\partial^2 N_i N_j (\phi_j - \phi_i) \Psi}{\partial N_j \partial N_i} + \frac{\partial^2 N_j (\phi_j - \phi_i) \Psi}{\partial N_j \partial \phi_i} + \frac{\partial^2 (1 - \phi_j) \phi_j (\phi_j - \phi_i) \Psi}{\partial \phi_j \partial \phi_i} \right\}$$

$$- D \sum_{ij} \Theta_{ij} \frac{\partial}{\partial \phi_i} (\phi_j - \phi_i) \Psi. \quad (125)$$

It can be checked immediately that (125) leads to $\int \prod_{k=1}^{N} (dN_k \, d\phi_k) \, \Psi = \text{const}$. Now we use (117), (124) and (122) and obtain the relation

$$\int \prod_{k=1}^{N} (dN_k \, d\phi_k) \Psi = \int \prod_{i=1}^{N} (d\bar{A}_i^+ \, d\bar{A}_i^-) \prod_{i=1}^{N} \left[ \bar{A}_i^+ + \bar{A}_i^- \right] P(\bar{A}_i, \tau; \bar{\nu}_0, 0)$$

$$= \left( \prod_{i=1}^{N} \left[ \bar{A}_i^+ + \bar{A}_i^- \right] \right) \text{const.} \quad (126)$$

Because of the fact that the integral $\int \prod_{k=1}^{N} (dN_k \, d\phi_k) \Psi$ is independent of time, the average $\langle \prod_{i=1}^{N} (\bar{A}_i^+ + \bar{A}_i^-) \rangle$ is also conserved. On the other hand, the initial state is always characterized
by $\overline{A}_i^+ + \overline{A}_i^- \equiv 1$, see also (112). Therefore, $\Psi$ is characterized by the normalization condition

$$\int \prod_{k=1}^N (dN_k \, d\phi_k) \Psi(N, \phi, t; N_0, \phi_0, 0) = 1.$$ \hspace{1cm} (127)

From this point of view, $\Psi$ has all properties of a probability distribution function or, because of its dependence on the initial conditions, of a conditional probability distribution function.

### 6.2.4. Reduced probability distribution.

Equation (123) can be written as

$$\left\langle \overline{A}_i^+ (t|\tilde{n}_i) \prod_{j \neq i}^N \left[ \overline{A}_j^+ (t|\tilde{n}_j) + \overline{A}_j^- (t|\tilde{n}_j) \right] \right\rangle = \int \prod_{k=1}^N d\phi_k \phi_i \left[ \int \prod_{k=1}^N dN_k \Psi(N, \phi, t; N_0, \phi_0, 0) \right].$$ \hspace{1cm} (128)

Obviously, the knowledge of the total probability distribution $\Psi(N, \phi, t; N_0, \phi_0, 0)$ is not necessary for the determination of averages and correlation functions. In principle, the reduced probability

$$\Psi(\phi, t; \phi_0, 0) = \left[ \int \prod_{k=1}^N dN_k \Psi(N, \phi, t; N_0, \phi_0, 0) \right] \hspace{1cm} (129)$$

is sufficient for the calculation of the above-mentioned statistical quantities. Note that each initial configuration $\tilde{n}_i$ is equivalent to $N_{i,0} \equiv 1$. The integration (129) is performed in each term of (125) under consideration of the usual boundary conditions $\Psi(N, \phi, t; N_0, \phi_0, 0) \to 0$ and $\partial_N \Psi(N, \phi, t; N_0, \phi_0, 0) \to 0$ for $\|N\| \to \infty$. This yields a Fokker–Planck equation for the reduced probability distribution function:

$$\frac{\partial}{\partial t} \Psi = \beta^\prime \sum_i \frac{\partial}{\partial \phi_i} \phi_i \Psi - \sum_{ij} \Theta_{ij} \frac{\partial}{\partial \phi_i} \left[ \tilde{\lambda} (1 - \phi_i) - \beta \phi_i \phi_j + D(\phi_j - \phi_i) \right] \Psi$$

$$+ \sum_{ij} \Theta_{ij} \frac{\partial^2}{\partial \phi_i \partial \phi_j} \left[ \tilde{\lambda} (1 - \phi_i) \phi_i (1 - \phi_j) - \beta (1 - \phi_i) \phi_i \phi_j - D(1 - \phi_i) \phi_j (\phi_j - \phi_i) \right] \Psi$$

$$+ \frac{z}{2} \sum_i \frac{\partial^2}{\partial \phi_i^2} \left[ \tilde{\lambda} (1 - \phi_i)^2 (1 + \phi_i^2) + \beta (2 \phi_i^2 - 2 \phi_i^3 + \phi_i^4) + D(\phi_i^2 + (1 - \phi_i)^2) \right] \Psi. \hspace{1cm} (130)$$

Obviously, it seems more correct to say that this final equation is a Fokker–Planck equation equivalent to the original master equations on a lattice with local complete exclusion rules. The new set of variables $\phi_i$ replaces the original discrete occupation numbers $n_i$. A first important remark concerns the mathematical structure of the new variables. While the occupation numbers are discrete quantities with only two values, $n_i = 0, 1$, the new variables $\phi_i$ may be continuous quantities, $\phi_i \in (-\infty, \infty)$. However, all moments and correlation functions of the original variables have an analogous representation in terms of the new variables.
6.3. Averages and correlation functions

A knowledge of the probability distribution function \( P(\phi_0, 0) \) allows the determination of all relevant averages and correlation functions by simple relations. Thus, the average \( \langle n_i(t) \rangle = \langle A_i^+(t) \rangle \) is given by (107) and (128), i.e.

\[
\langle A_i^+(t) \rangle = \sum_{\phi_0} \int \prod_{k=1}^N d\phi_k \phi_i \Psi(\phi, t; \phi_0, 0) P(\phi_0, 0) = \langle \phi_i \rangle .
\]

The connection between the initial probabilities \( P(\phi_0, 0) \) and \( P(n', 0) \) is given by the simple relation \( \phi_i, \phi_{i,0} = n_i' \). Thus the average \( \langle A_i^+(t) \rangle \) (determined from the Fock space representation and the original master equation) and the average of \( \phi_i \) using the statistical weight \( \Psi \) are equivalent observables. Higher moments are given by similar relations, e.g.

\[
\langle A_i^+(t)A_j^+(t) \rangle = \sum_{\phi_0} \int \prod_{k=1}^N d\phi_k \phi_i \phi_j \Psi(\phi, t; \phi_0, 0) P(\phi_0, 0) = \langle \phi_i(t)\phi_j(0) \rangle .
\]

Furthermore, correlation functions can be determined by using (115) and (134):

\[
\langle A_i^+(t)A_j^+(0) \rangle = \sum_{\phi_0} \int \prod_{k=1}^N d\phi_k \phi_i \Psi(\phi, t; \phi_0, 0) \phi_j P(\phi_0, 0) = \langle \phi_i(t)\phi_j(0) \rangle .
\]

A further test of these relations is demonstrated in appendix C. On the other hand, it is simple to demonstrate that \( \langle \phi_i^2 \rangle \neq \langle n_i^2 \rangle = \langle n_i \rangle \). This is a consequence of the anticommutation algebra, which was used for the derivation of (130). This algebra excludes multinomials containing variables with a power higher than one. Obviously, this discussion together with some general considerations, suggest that all averages over multilinear combinations \( \langle \phi_i\phi_j\phi_k \cdots \phi_m \rangle \) are equivalent to the corresponding averages \( \langle A_i^+ A_j^+ A_k^+ \cdots A_m^+ \rangle \) = \( \langle n_i n_j n_k \cdots n_m \rangle \), while all averages of multinomials with a higher degree per variable, e.g. \( \langle \phi_i^2 \phi_j \phi_k \cdots \phi_m \rangle \), have no direct physical meaning. A similar statement follows for the two-time correlation function \( \langle A_i^+(t)A_j^+(t) \cdots A_i^+(0)A_j^+(0) \cdots \rangle \) if the multinomials corresponding to the time \( t \) and to the initial time are linear in their variables.

7. Discussion

The derivation of a Fokker–Planck equation describing a substitute process instead of the original process is a particularly elegant technique for the analysis of chemical master equations. Without the exclusion principle, a well-established method is the so-called Poisson representation [41, 42]. Here, it was assumed that the probability distribution function \( P(n, t) \) of a lattice is a superposition of multivariate uncorrelated Poisson processes

\[
P(n, t) = \int d\theta \prod \frac{\exp[-\theta_i] \theta_i^{n_i}}{n_i!} f(\theta, t) .
\]

In contrast to processes under exclusion rules, the occupation numbers are non-negative integers, \( n_i = 0, 1, 2, \ldots \). Elementary diffusion steps are defined between neighbour lattice sites, whereas
chemical reactions occur between the particles of one site. Thus, chemical reactions are totally local processes, while the elementary diffusion processes require two lattice sites. For our reaction–diffusion system, the function $f(\theta, t)$ defined in (134) satisfies the equation

$$\frac{\partial f(\theta, t)}{\partial t} = -D \sum_{ij} \Theta_{ij} \frac{\partial}{\partial \theta_i} (\theta_j - \theta_i) f(\theta, t) - \sum_i \frac{\partial}{\partial \theta_i} \left[ (\tilde{\lambda} - \beta') \theta_i - \beta \theta_i^2 \right] f(\theta, t)$$

$$+ \sum_i \frac{\partial^2}{\partial \theta_i^2} (\tilde{\lambda} \theta_i - \beta \theta_i^2) f(\theta, t),$$

(135)

which is of a Fokker–Planck form provided $\tilde{\lambda} \theta_i > \beta \theta_i^2$. There exists a simple relationship between the moments of the occupation numbers and moments formed by application of $f(\theta, t)$

$$\langle n_i n_j \cdots n_m \rangle = \langle \theta_i \theta_j \cdots \theta_m \rangle = \int d\theta \langle \theta_i \theta_j \cdots \theta_m \rangle f(\theta, t).$$

(136)

This relation follows from the factorial moments of the Poisson distribution. However, $f(\theta, t)$ is not a real probability distribution function because it cannot be guaranteed that this pseudo-probability distribution is a positive-definite function. But it can demonstrated that (141) is equivalent to a stochastic differential equation

$$\frac{d\theta_i}{dt} = (\tilde{\lambda} - \beta') \theta_i - \beta \theta_i^2 + D \sum_j \Theta_{ij} (\theta_j - \theta_i) + \xi_i(t)$$

(137)

with

$$\langle \xi_i(t) \xi_j(t') \rangle = \delta_{ij} [\tilde{\lambda} \theta_i - \beta \theta_i^2] \delta(t - t').$$

(138)

A more detailed analysis shows that the motion takes place over the range $(0, \tilde{\lambda} / \beta)$ and both boundaries satisfy the criteria for entrance boundaries. This means, if the initial conditions are located on this range, the system cannot leave this range. The time-dependent functions $\theta_i(t)$ define an underlying substitute process.

The corresponding substitute process for the diffusion reaction system with exclusion rules reads

$$\frac{d\phi_i}{dr} = -\beta' \phi_i + \sum_j \Theta_{ij} [\tilde{\lambda} \phi_j (1 - \phi_i) - \beta \phi_i \phi_j + D(\phi_j - \phi_i)] + \xi_i(t),$$

(139)

with the noise $\xi_i(t)$ defined by

$$\langle \xi_i(t) \xi_j(t') \rangle = \Theta_{ij} \left[ \tilde{\lambda} (1 - \phi_i) \phi_j (1 - \phi_j) - \beta (1 - \phi_i) \phi_i \phi_j - D (1 - \phi_i) \phi_j (\phi_j - \phi_i) \right]$$

$$+ \frac{z}{2} \delta_{ij} \left[ \tilde{\lambda} (1 - \phi_i)^2 (1 + \phi_j^2) + \beta (2 \phi_i^2 - 2 \phi_i^3 + \phi_j^4) + D \left[ \phi_i^2 + (1 - \phi_i)^2 \right]^2 \right].$$

(140)

Both substitute processes with and without exclusion rules become equivalent for purely unimolecular reactions. In our case, this means $D = \tilde{\lambda} = \beta = 0$, and we get the deterministic equations

$$\frac{d\theta_i}{dt} = -\beta' \theta_i \quad \text{and} \quad \frac{d\phi_i}{dr} = -\beta' \phi_i$$

(141)
describing the reaction $A \rightarrow 0$. Surprisingly, these processes are not controlled by noise terms, although the corresponding real evolutions of the local occupation number are random processes. This is a typical indication that (137) and (139) describe substitute processes.

However, there are essential differences between the substitute processes with and without exclusion rules. The first difference concerns the diffusion process. While the Poisson representation describes diffusion by a deterministic equation

$$\frac{d\theta_i}{dt} = D \sum_j \Theta_{ij}(\theta_j - \theta_i), \quad (142)$$

the substitute process with exclusion requires an additional noise term

$$\frac{d\phi_i}{dt} = D \sum_j \Theta_{ij}(\phi_j - \phi_i) + \xi_i(t). \quad (143)$$

The origin of this surprising difference comes from the vacancies. The exclusion principle requires that an arbitrary site is either occupied or vacant. Without the exclusion principle, this conservation law is violated since the basin of vacancies per lattice site is infinitely large.

A second difference between both approaches occurs in the deterministic bimolecular reaction part. The exclusion principle allows only reaction terms connecting neighboured lattice sites. In contrast to this behaviour, the bimolecular reaction terms may be local terms if the exclusion principle is neglected. Therefore, the corresponding noise without the exclusion principle may also be a local expression, while the noise term for bimolecular reactions with exclusion rules are necessarily nonlocal.

Another important difference between both representations is the range of allowed values. As mentioned above, the variables of (137) have to be localized over the range $0 < \theta_i < \tilde{\lambda}/\beta$. On the other hand, each well-defined initial value $\theta_i(0) = \theta_i^0$ corresponding to an initial distribution $f(\theta_i, 0) = \delta(\theta_i - \theta_i^0)$ is related to a reasonable probability distribution of the occupation number

$$P(n_i, 0) = \exp\left(-\theta_i^0\right) \left(\theta_i^0\right)^{n_i} / n_i! \quad (144)$$

Thus, an initial value $\theta_i^0 > \tilde{\lambda}/\beta$ should also be allowed. This means that the correlation of the noise terms becomes negative and, consequently, (135) is no longer a Fokker–Planck equation. On the other hand, the process (139) allows the full range $-\infty < \phi_i < \infty$ for all variables. Especially, the corresponding equation (130) is a true Fokker–Planck equation in the sense that the kernel of the second-order derivatives is positive definite. This may be checked by the fact that (114) is always a true Fokker–Planck equation with a positive-definite second-order kernel. Thus, during all the following transformations of the probability distribution function and the variables, this characteristic property of (114) remains unchanged. In particular, we get the result that the noise matrix (140) is also a positive-definite object, so that (130) for the substitute process (139) is defined for all possible realizations of the variables $\phi$.

We remark that another approach leading to a field theoretical description [43] of reaction–diffusion systems considering diffusive, reactive and branching processes of order higher than $n = 2$ also use the Fock space representation and the path-integral technique. But the main difference from our approach is that the original master equation is transformed into a Fock space.
representation by the application of a bosonic algebra with the consequence that the exclusion principle is completely neglected. In contrast, the fermionic algebra used in the present approach guarantees the exclusion principle, but it becomes very difficult to construct a treatable path integral for reactive processes of orders $n > 2$.

Finally, we compare the classical evolution equation (10) for a reaction–diffusion system with the coarse-grained versions of the substitute processes (137) and (139). A simple method is a kind of random-phase approximation. Here, we neglect all local fluctuations. Thus we may interpret $\phi_i$ as a lattice representation of the local particle density $\rho(r, t)$. If the spatial fluctuations of $\rho(r, t)$ are sufficiently small, we arrive at

$$\frac{d\rho}{dt} = +D'\Delta\rho + \left( z\bar{\lambda} - \beta' \right)\rho - z(\bar{\lambda} + \beta)\rho^2 + \zeta(r, t)$$

with the noise

$$\langle \zeta(r, t)\zeta(r', t') \rangle \sim \rho(r, t)\delta(r - r')\delta(t - t').$$

Note that the diffusion coefficient $D' = D\ell^2$ contains now also information about the characteristic length scale $\ell$ of the lattice. Thus, the kinetic equation (10) may be interpreted as the coarse-grained version of the stochastic evolution equation (139).

8. Conclusions

It was shown that master equations of discrete variables describing diffusion processes as well as unimolecular and bimolecular reaction processes on a lattice can be mapped onto a system of Ito stochastic differential equations. These stochastic equations are characterized by the following special properties:

(i) All equations appear to be local processes (i.e. processes with respect to one lattice cell or lattice point). Thus, each lattice site $i$ corresponds to a closed system of differential equations consisting of dynamical variables ($A^+_i$ and $A^-_i$), which are related only to this lattice site. In other words, the differential equations are separated with respect to the dynamical variables of each lattice site, see (102).

(ii) The dynamic observables of different lattice sites only seem to be independent. The coupling between different lattice points is a result of the simultaneous action of Wiener processes $W_{a}(t) \leftrightarrow \phi_{ij}(t), \psi_{ij}(t), \omega_{ij}(t), \ldots$ at pairs of lattice sites.

Thus, only the stochastic processes enforce the kinetic interaction between neighbouring sites in this representation. Since the Ito stochastic differential equations are equivalent to a Fokker–Planck equation, one obtains after some reasonable transformations a probability distribution, which allows the determination of averages, moments and correlation functions by formulae similar to the determination of these statistical variables by the original master equation, see (131)–(133). The decisive difference between the original probability distribution function (corresponding to the master equation) and the new probability distribution related to a Fokker–Planck equation is given by the variables. The master equation of the original diffusion reaction process on the lattice is based on a finite number of discrete variables, while the probability distribution function of the obtained substitute process depends on continuous variables (in the present case $\phi_i$). This mapping from a discrete representation to a continuous one is the main result of the present work.
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Appendix A. Infinitesimal transition operator

We assume that \( \hat{E}'(t) dt \) and \( \hat{E}(t) dt \), see (82) and (86), have similar mathematical structures, i.e. we may write

\[
\hat{E}'(t) dt = \hat{a}^\dagger M_0 \hat{a} dt + \sum_{\alpha=1}^{N_W} (\hat{a}^\dagger \Xi^{\alpha} \hat{a}) dW_\alpha(t) + \gamma' \hat{a}_1^\dagger \hat{a}_1 \hat{a}_2^\dagger \hat{a}_2 dt.
\]  

(A.1)

Thus, the expansion of \( \exp[\hat{E}'(t) dt] \) in powers of \( dt \) up to the second order (higher orders are not considered, because of equation (81)) leads to

\[
\exp[\hat{E}'(t) dt] = 1 + \hat{E}'(t) dt + \frac{1}{2} [\hat{E}'(t) dt]^2.
\]  

(A.2)

Considering (81), the last term of (A.2) can be rewritten as

\[
[\hat{E}'(t) dt]^2 = \sum_{\alpha=1}^{N_W} (\hat{a}^\dagger \Xi^{\alpha} \hat{a}) dW_\alpha(t) \sum_{\beta=1}^{N_W} (\hat{a}^\dagger \Xi^{\beta} \hat{a}) dW_\beta(t)
\]

\[
= \sum_{\alpha, \beta} (\hat{a}^\dagger \Xi^{\alpha} \hat{a})(\hat{a}^\dagger \Xi^{\beta} \hat{a}) dW_\alpha(t) dW_\beta(t)
\]

\[
= \sum_{\alpha, \alpha'} \sum_{ab,cd} (\Xi_{ab}^{\alpha} \Xi_{cd}^{\alpha'}) \hat{a}_{a}^\dagger \hat{a}_{b}^\dagger \hat{a}_{d} \hat{a}_{d} dt
\]

\[
= \sum_{\alpha, \alpha'} \sum_{ab,cd} (\Xi_{ab}^{\alpha} \Xi_{cd}^{\alpha'}) \hat{a}_{a}^\dagger (\delta_{cb} - \hat{a}_{c}^\dagger \hat{a}_{b}) \hat{a}_{d} dt.
\]  

(A.3)

The remaining fourth-order term becomes after some simple transformations

\[
- \sum_{\alpha} \sum_{ab,cd} (\Xi_{ab}^{\alpha} \Xi_{cd}^{\alpha'}) \hat{a}_{a}^\dagger \hat{a}_{b}^\dagger \hat{a}_{d} \hat{a}_{d} = -2 \sum_{\alpha} (\Xi_{11}^{\alpha} \Xi_{22}^{\alpha}) \hat{a}_{1}^\dagger \hat{a}_{2}^\dagger \hat{a}_{1} \hat{a}_{2} - 2 \sum_{\alpha} (\Xi_{12}^{\alpha} \Xi_{21}^{\alpha}) \hat{a}_{1}^\dagger \hat{a}_{2}^\dagger \hat{a}_{1} \hat{a}_{2}
\]

\[
= 2 \sum_{\alpha} (\Xi_{11}^{\alpha} \Xi_{22}^{\alpha}) \hat{a}_{1}^\dagger \hat{a}_{2}^\dagger \hat{a}_{2} - 2 \sum_{\alpha} (\Xi_{12}^{\alpha} \Xi_{21}^{\alpha}) \hat{a}_{1}^\dagger \hat{a}_{2}^\dagger \hat{a}_{2}
\]

\[
= 2 \sum_{\alpha} \det(\Xi^{\alpha}) \hat{a}_{1}^\dagger \hat{a}_{1} \hat{a}_{2}^\dagger \hat{a}_{2}.
\]  

(A.4)

Hence,

\[
[\hat{E}'(t) dt]^2 = \left[ \sum_{\alpha} \sum_{a b d} (\Xi_{a b}^{\alpha} \Xi_{b d}^{\alpha'}) \hat{a}_{a}^\dagger \hat{a}_{d} + 2 \sum_{\alpha} \det(\Xi^{\alpha}) \hat{a}_{1}^\dagger \hat{a}_{1} \hat{a}_{2}^\dagger \hat{a}_{2} \right] dt.
\]  

(A.5)

The substitution of (A.5) into (A.2) and the comparison with (86) yield

\[
\Xi^{\alpha} = \Xi^{\alpha'}, \quad \gamma = \gamma' + \sum_{\alpha} \det(\Xi^{\alpha}), \quad M_0 = M_0' + \frac{1}{2} \sum_{\alpha} \Xi^{\alpha} \xi^{\alpha}.
\]
Appendix B. Grassmann representation of Ito stochastic differential equations

We derive here the representation of an Ito differential equation for an infinitesimal time interval. It follows from (85) and (89) that

\[ |F(dt)⟩ = \exp{\{ \hat{E}'(t) \, dt \}}|n'⟩ = \exp{\{ \hat{E}'(t) \, dt \}}(\bar{\alpha}_1)^{n'}(\bar{\alpha}_2)^{1-n'}. \]  
\[ \text{(B.1)} \]

The introduction of a $\delta$ function leads to

\[ |F(dt)⟩ = \int \exp{\{ \hat{E}'(t) \, dt \}} \prod_{a=1}^{2} [\, d\bar{\alpha}_a^0 \, d\alpha_a^0 \exp\{\alpha_a^0(\bar{\alpha}_a^0 - \bar{\alpha}_a^1)\} (\bar{\alpha}_1^0)^{n'}(\bar{\alpha}_2^0)^{1-n'}. \]  
\[ \text{(B.2)} \]

Using the Grassmann integral representation of the $\delta$ function, (B.2) can be rewritten as

\[ |F(dt)⟩ = \int \exp{\{ \hat{E}'(t) \, dt \}} \prod_{a=1}^{2} [\, d\alpha_a^0 \, d\bar{\alpha}_a^0 \exp\{\alpha_a^0(\bar{\alpha}_a^0 - \bar{\alpha}_a^1)\} (\bar{\alpha}_1^0)^{n'}(\bar{\alpha}_2^0)^{1-n'}. \]  
\[ \text{(B.3)} \]

The expansion of $\exp{\{ \hat{E}'(t) \, dt \}}$ in powers of $dt$ leads to (86), see also appendix A; only the Fermi operators must be replaced by the corresponding Grassmann variables and Grassmann differential operators. One obtains the result:

\[ \exp{\{ \hat{E}'(t) \, dt \}} = 1 + \sum_{a,b} \bar{\alpha}_a [\, M_{0}^{ab} \, dt + \sum_{a} \xi_{ab}^a \, dW_a(t) \]  
\[ \frac{\partial}{\partial \bar{\alpha}_b} + \gamma \, dt \bar{\alpha}_1 \frac{\partial}{\partial \bar{\alpha}_1} \frac{\partial}{\partial \bar{\alpha}_2}. \]  
\[ \text{(B.4)} \]

The consideration of

\[ \frac{\partial}{\partial \bar{\alpha}_b} \exp\left\{ - \sum_{a=1}^{2} \alpha_a^0 \bar{\alpha}_a \right\} = \alpha_b^0 \exp\left\{ \sum_{a=1}^{2} -\alpha_a^0 \bar{\alpha}_a \right\} \]  
\[ \text{(B.5)} \]

yields:

\[ \exp{\{ \hat{E}'(t) \, dt \}} \exp\left\{ - \sum_{a=1}^{2} \alpha_a^0 \bar{\alpha}_a \right\} \]  
\[ = \left( 1 + \sum_{a,b} \bar{\alpha}_a [\, M_{0}^{ab} \, dt + \sum_{a} \xi_{ab}^a \, dW_a(t) \]  
\[ \alpha_b^0 + \gamma \, dt \bar{\alpha}_1 \alpha_1^0 \bar{\alpha}_2 \alpha_2^0 \right) \exp\left\{ - \sum_{a=1}^{2} \alpha_a^0 \bar{\alpha}_a \right\}. \]  
\[ \text{(B.6)} \]

The right-hand side should be represented again by an exponential function, i.e. we need a solution of

\[ 1 + \sum_{a,b} \bar{\alpha}_a [\, M_{0}^{ab} \, dt + \sum_{a} \xi_{ab}^a \, dW_a(t) \]  
\[ \alpha_b^0 + \gamma \, dt \bar{\alpha}_1 \alpha_1^0 \bar{\alpha}_2 \alpha_2^0 = \exp{\{ E''(t, \bar{\alpha}, \alpha^0) \, dt \}. \]  
\[ \text{(B.7)} \]
Appendix A suggests the following structure of \( E''(t, \alpha, \alpha_0) \):

\[
E''(t, \alpha, \alpha_0) \,dt = \sum_{a,b} a_{\alpha} \left[ M_0^a_{ab} \,dt + \sum_\alpha \Xi''_{\alpha a} \,dW_{\alpha}(t) \right] \alpha^0_b + \gamma' \,dt \alpha^0_1 \alpha^0_2. \tag{B.8}
\]

The expansion of \( \exp\{E''(t, \alpha, \alpha_0) \,dt\} \) in powers of \( dt \) leads to

\[
\exp \{E''(t, \alpha, \alpha_0) \,dt\} = 1 + E''(t, \alpha, \alpha_0) \,dt + \sum_\alpha \det |\Xi''_{\alpha a}| dt \alpha^0_1 \alpha^0_2. \tag{B.9}
\]

Note that the algebraic procedure is similar to appendix A. A comparison between (B.6) and (B.9) shows the identities:

\[
M_0 = M_0', \quad \Xi'' = \Xi^{''\alpha} \quad \text{and} \quad \gamma = \gamma'' + \sum_\alpha \det |\Xi''_{\alpha a}|. \tag{B.10}
\]

The matrices \( M_0 \) and \( \Xi'' \) are completely defined by the Ito stochastic differential equation (83). On the other hand, the value of the quantity \( \gamma \) can be arbitrarily chosen because \( \hat{C}_1 \hat{a}_1 \hat{a}_2 \) vanishes identically in the reduced Fock space. Thus, one obtains the possibility for a calibration. We define the gauge measure by setting \( \gamma'' = 0 \), i.e. the original evolution operator (68), see also (82), \( \hat{E}(t) \) must be replaced by

\[
\hat{\hat{E}}(t) \rightarrow \hat{\hat{E}}(t) - \sum_\alpha \det |\Xi''_{\alpha a}| \,dt \alpha^0_1 \alpha^0_2. \tag{B.11}
\]

This operator corresponds to the evolution function \( E''(t, \alpha, \alpha_0) \) defined by Grassmann variables:

\[
E''(t, \alpha, \alpha_0) \,dt = \sum_{a,b} a_{\alpha} \left[ M_0''_{ab} \,dt + \sum_\alpha \Xi''_{\alpha a} \,dW_{\alpha}(t) \right] \alpha^0_b. \tag{B.12}
\]

The origin of this degree of freedom is the existence of more than one zero operator, i.e. we have several operators with the property \( \hat{C}|n\rangle = 0 \) independent of the actual state \( |n\rangle \). The complete Fock space \( \mathcal{F} \) has only the trivial zero operator \( \hat{0} \), but the subspace \( \mathcal{F}_0 \) is related to a large class of zero operators, see (41). This property will be transferred onto the mathematical structure of the Grassmann representation and is related to the free calibration of \( \gamma'' \).

However, any Ito stochastic differential equation (83) can be mapped onto an evolution function (B.8) defined by Grassmann variables.

**Appendix C. Hierarchical evolution equations**

We demonstrate that the evolution equations for averages and second moments are equivalent for both the Fock space representation and the Fokker–Planck equation (130) for the pure autocatalytic reaction. In a way, these identities can be understood as a further test of the validity of (130).
C.1. Hierarchical equations in the Fock space

The evolution equation for the average $\overline{A}^+_k(t)$ follows immediately from the general equations (13) and (37):

$$\frac{\partial}{\partial t} \overline{A}^+_k(t) = \frac{\partial}{\partial t} \langle s| \hat{A}^+_k | F(t) \rangle = \langle s| [\hat{A}^+_k, \hat{L}] | F(t) \rangle + \langle s| \hat{L} \hat{A}^+_k | F(t) \rangle. \tag{C.1}$$

The second term vanishes identically because of the normalization condition $\langle s| \hat{L} = 0$. Considering (29), (30) and (39), it follows for the autocatalytic process

$$\hat{L} = \sum_{i,j} \Theta_{ij} \hat{a}^+_i \hat{a}_j \hat{L}_i = \sum_{i,j} \Theta_{ij} \hat{L}_i \hat{A}^+_j \tag{C.2}$$

and therefore

$$\langle s| [\hat{A}^+_k, \hat{L}] | F(t) \rangle = \sum_{i,j} \Theta_{ij} \langle s| [\hat{A}^+_k, \hat{L}_i] \hat{A}^+_j | F(t) \rangle = \sum_{j} \Theta_{kj} \langle s| [\hat{A}^+_k, \hat{L}_k] \hat{A}^+_j | F(t) \rangle. \tag{C.3}$$

Using (27) and (39), the evaluation of the commutator $[\hat{A}^+_k, \hat{L}_k]$ yields

$$\frac{\partial}{\partial t} \overline{A}^+_k = \sum_{j} \Theta_{kj} [\lambda \overline{A}^+_j, \overline{A}^+_j] - (\lambda + \beta) \langle \hat{A}^+_k \hat{A}^+_j \rangle. \tag{C.4}$$

An analogous procedure leads to the evolution equation for the second moment:

$$\frac{\partial}{\partial t} \langle \hat{A}^+_k \hat{A}^+_k \rangle = \Theta_{kl} [\lambda (\overline{A}^+_k + \overline{A}^+_l) - 2(\lambda + \beta) \langle \hat{A}^+_k \hat{A}^+_l \rangle]
+ \sum_{j} \Theta_{kj} (1 - \delta_{lj}) [\lambda \langle \hat{A}^+_k \hat{A}^+_j \rangle - (\lambda + \beta) \langle \hat{A}^+_k \hat{A}^+_j \hat{A}^+_j \rangle]
+ \sum_{j} \Theta_{lj} (1 - \delta_{kj}) [\lambda \langle \hat{A}^+_k \hat{A}^+_j \rangle - (\lambda + \beta) \langle \hat{A}^+_k \hat{A}^+_j \hat{A}^+_j \rangle]. \tag{C.5}$$

C.2. Hierarchical equations from Fokker–Planck equations

The evolution of the average $\langle \phi_k \rangle$ can be obtained from (131):

$$\frac{\partial}{\partial t} \langle \phi_k \rangle = \sum_{\phi_0} \int \prod_{n=1}^{N} d\phi_n \phi_k \left\{ \frac{\partial}{\partial t} \Psi (\phi, t; \phi_0, 0) \right\} P(\phi_0, 0). \tag{C.6}$$

Using (136), the evolution equation becomes

$$\frac{\partial}{\partial t} \langle \phi_k \rangle = \frac{z}{2} \sum_{\phi} \sum_{\phi_i} \int \prod_{n=1}^{N} d\phi_n \phi_i \frac{\partial^2}{\partial \phi_i^2} \left\{ (\lambda + \beta) (1 - \phi_i)^2 \phi_i^2 + \lambda (1 - \phi_i)^2 + \beta \phi_i^2 \right\} \times \Psi (\phi, t; \phi_0, 0) P(\phi_0, 0)$$

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\[ + \sum_{ij} \Theta_{ij} \sum_{\phi_0} \prod_{n=1}^{N} d\phi_n \phi_i \frac{\partial^2}{\partial \phi_i \partial \phi_j} [\lambda (1 - \phi_i) - \beta \phi_i] (1 - \phi_j) \phi_j \]
\[ \times \Psi(\phi, t; \phi_0, 0) P(\phi_0, 0) \]
\[ + \sum_{ij} \Theta_{ij} \sum_{\phi_0} \prod_{n=1}^{N} d\phi_n \phi_k \frac{\partial}{\partial \phi_j} [\beta \phi_i - \lambda (1 - \phi_i)] \phi_j \Psi(\phi, t; \phi_0, 0) P(\phi_0, 0). \quad (C.7) \]

Partial integration considering the boundary conditions \( \Psi(\phi, t; \phi_0, 0) \rightarrow 0 \) for \( \| \phi \| \rightarrow \infty \) leads to the equation
\[ \frac{\partial}{\partial t} \langle \phi_k \rangle = \sum_j \Theta_{kj} \{ \lambda \langle \phi_j \rangle - (\lambda + \beta) \langle \phi_k \phi_j \rangle \}. \quad (C.8) \]

An analogous procedure using (138) and (136) yields
\[ \frac{\partial}{\partial t} \langle \phi_k \phi_j \rangle = \Theta_{kj} \{ \lambda (\langle \phi_k \rangle + \langle \phi_j \rangle) - 2(\lambda + \beta) \langle \phi_k \phi_j \rangle \}
\[ + \sum_j \Theta_{kj} (1 - \delta_{ij}) \{ \lambda \langle \phi_i \phi_j \rangle - (\lambda + \beta) \langle \phi_k \phi_i \phi_j \rangle \}
\[ + \sum_j \Theta_{ij} (1 - \delta_{kj}) \{ \lambda \langle \phi_k \phi_j \rangle - (\lambda + \beta) \langle \phi_k \phi_i \phi_j \rangle \}. \quad (C.9) \]

The comparison of (C.4) with (C.8) and (C.5) with (C.9) shows a complete correspondence. This agreement confirms the equivalence between the original master equation of the autocatalytic reaction processes and the Fokker–Planck equation (130).

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