Langevin Equations for Reaction-Diffusion Processes

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For reaction-diffusion processes with at most bimolecular reactants, we derive well-behaved, numerically tractable, exact Langevin equations that govern a stochastic variable related to the response field in field theory. Using duality relations, we show how the particle number and other quantities of interest can be computed. Our work clarifies long-standing conceptual issues encountered in field-theoretical approaches and paves the way for systematic numerical and theoretical analyses of reaction-diffusion problems.

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An important challenge in many areas of science is to reliably derive Langevin equations (LEs) governing the dynamics of relevant degrees of freedom at mesoscopic or coarse-grained scales. LEs are not only useful numerically, saving one from dealing with unessential details, but they also constitute the usual starting point for analytic approaches such as renormalization group analyses [1]. Not surprisingly, there is no systematic method to derive LEs from first principles, given that they are supposed to resum the complicated effects of different "forces" acting on the system at microscopic scales. As a matter of fact, LEs are often built from the underlying "mean-field" deterministic dynamics to which a noise term is added (see, e.g., [2,3]). Several attempts to go beyond such a heuristic approach have been made. Most use more or less sophisticated approximations to derive an equation for a "density" field [4–7]. The most popular ones are van Kampen's system size expansion [8] and Gillespie's projection method [9] (equivalent to a second-order truncation of the Kramers-Moyal equation), both of which rely on the presence of large numbers of particles. Although powerful within their range of validity, these approximations fail for spatially extended systems displaying empty or sparsely populated regions.

Reaction-diffusion (RD) processes, which represent a wealth of phenomena in physics, chemistry, biology, population genetics, and even linguistics [10–13], play a particular role in this context in two respects: (i) they often exhibit empty domains and transition to absorbing states with strong fluctuations, and thus stand out of the validity range of popular methods to write LEs; (ii) their microscopic dynamics can be described exactly by a master equation, an object that can only be used directly for small systems but constitutes the starting point of field-theoretical approaches with which, in turn, LEs can be formally associated. This procedure, using the Doi-Peliti formalism [14–20], does not require large numbers of particles, and it would then appear that reliable LEs can be written for RD processes even in sparse regimes. However, as we show below, the LEs obtained often appear "paradoxical," the fields they govern may be difficult to interpret, and, worse, the path followed to derive them suffers from fundamental difficulties. Indeed, it often involves illegitimate steps, a consequence of which can be the advent of imaginary noise in equations supposed to describe real fields [21,22]. A famous example of these inconsistencies is the simple, yet puzzling, pure annihilation case where particles from a single species diffuse and annihilate by pair upon encounter [19,20,22–25].

In this Letter, changing perspective, we present a systematic and exact derivation of LEs for RD processes that does not rely on field-theoretic tools, but rather stems directly from the master equation. These LEs do *not* govern a density field, but, using duality considerations, a somewhat standard technique for stochastic processes and interacting particle systems [26–33], we show how the particle number and all its moments and correlations are computable from them. We also show how to relate our LEs to field-theoretical approaches, resolving all previous paradoxes, and clarifying the meaning of the stochastic variable they govern. On top of the resolution of these long-standing conceptual issues, we show that this approach provides us with new practical and theoretical tools for computing observables of RD processes.

We consider the class of single-species RD processes involving all possible reactions of the form $A \xrightarrow{\alpha_p} pA$ (p arbitrary, combined with any mixture of pairwise annihilation-coagulation $2A \xrightarrow{\beta_p} \emptyset$ and/or A (with respective rates β_0, β_1). This set of reactions—comprising at most bimolecular reactants—already encompasses most of the physically interesting cases. For the sake of notational simplicity, we deal with the zero-dimensional case in what follows, because by considering diffusion as the nearest-neighbor hopping reaction $A\emptyset \xleftrightarrow{D} \emptyset A$, the generalization to d dimensions is straightforward (see [34] for a detailed treatment). We define $N(t)$ as the stochastic variable representing the number of particles in the system at time t. The master equation that describes the evolution of the probabilities $P_n(t) \equiv \mathrm{Prob}[N(t) = n]$ reads

$$\partial_t P_n(t) = \sum_m L_{nm} P_m(t), \tag{1}$$

where the elements of the rate transition matrix L are $L_{nm} = \sum_p \alpha_p m(\delta_{n+1-p,m} - \delta_{m,n}) + \sum_q \beta_q m(m-1)(\delta_{n+2-q,m} - \delta_{m,n})$. Introducing the probability-generating function $G(z,t) = \sum_{n=0}^{\infty} z^n P_n(t)$, Eq. (1) can be subsumed as an evolution equation for $G(z,t)$,

$$\partial_t G(z,t) = \mathcal{L}_z G(z,t) \tag{2}$$

which involves a second-order evolution operator $\mathcal{L}_z = \mathcal{L}(z, \partial_z) = A(z)\partial_z + B(z)\partial_z^2$, determined by the reaction rates through

$$A(z) = \sum_p \alpha_p (z^p - z), \qquad B(z) = \sum_q \beta_q (z^q - z^2). \tag{3}$$

Note that for the reactions we deal with, the coefficient of the second derivative reduces to a second-order polynomial in z,

$$B(z) = (\beta_0 + \beta_1)(z - \ell)(1 - z), \quad \ell \equiv -\beta_0/(\beta_0 + \beta_1). \tag{4}$$

The consideration of G—a well-defined analytic function for z in the complex unit disk $|z| \leq 1$—is a somewhat standard tool [35]. Besides the probability normalization encoded through $G(z = 1, t) = 1$, derivatives of $G(z,t)$ with respect to z evaluated at $z = 1$ give access (in principle) to the (factorial) moments of the number of particles. Some recent works in the literature have also shown how one can extract rare-events statistics from the behavior of $G(z,t)$ near $z = 0$ [36].

The second-order operator $\mathcal{L}_z$ governing the evolution of G is *not* a Fokker-Planck operator that could be associated with the number of particles $N(t)$. However, $\mathcal{L}_z^\dagger$, the Hermitian conjugate of $\mathcal{L}_z$, is the Fokker-Planck operator for a stochastic variable $Z(t)$ obeying the Itō (prepoint) LE,

$$\dot{Z}(t) = A(Z(t)) + \sqrt{2B(Z(t))}\eta(t), \tag{5}$$

where $\eta(t)$ is a zero-mean unit variance Gaussian white noise. Crucially, due to the form of the drift and diffusion functions $A(z)$, $B(z)$ given in Eqs. (3) and (4), the stochastic variable $Z(t)$ always remains (if initially so) in the bounded real interval $[\ell, 1]$ where $\sqrt{B(z)} \geq 0$. We show in the following that the determination of the statistics of $Z(t)$ through the knowledge of G suffices to extract all the physics of interest for the original RD process.

With its multiplicative noise and its two square-root barriers at ℓ and 1, Eq. (5) resembles phenomenological LEs usually considered for nonequilibrium phase transitions with symmetric absorbing states [37–43]. However, the LE (5) is exact and $Z(t)$ is *not* a density: we show below that $Z(t)$ is in fact closely related to the (time-reversed) response field used in field theory and explain how useful quantities can be computed from Eq. (5).

The evolution of $Z(t)$ stops when it has reached the absorbing barrier located at 1 (whose fixed location can be traced back to the probability conservation). This implies the existence of a delta-peak term at $z = 1$ in the probability distribution $p(z,t)$ of $Z(t)$ and, depending on the values of the α_p's, whenever (the otherwise always non-negative) $A(\ell)$ vanishes, a second delta peak appears at $z = \ell$. Thus, the general form of $p(z,t)$ reads

$$p(z,t) = p_c(z,t) + q_1(t)\delta(1 - z) + q_\ell(t)\delta(\ell - z), \tag{6}$$

where $p_c(z,t)$ is the continuous part of the distribution and q_1, q_ℓ the weights at the boundaries. Note, finally, that efficient and accurate methods dealing properly with multiplicative square-root noise exist [37,38,44–46], so that LE (5) can be used numerically (even in the spatially extended case), and thereby the obtained statistics of $Z(t)$ faithfully reconstructs Eq. (6).

We now derive the fundamental relation that allows us to compute quantities for the original RD process. Using Itō calculus in Eq. (5) [30,35] or, alternatively, its associated Fokker-Planck equation, one can show that

$$\partial_t \langle Z(t)^n \rangle_{\mathrm{LE}} = \sum_m (L^T)_{nm} \langle Z(t)^m \rangle_{\mathrm{LE}}, \tag{7}$$

where L^T is the transpose matrix of L [see Eq. (1)], and $\langle \cdot \rangle_{\mathrm{LE}}$ denotes averaging over the noise $\eta(t)$ of Eq. (5), that is, with the distribution $p(z,t)$. Combining Eqs. (1) and (7), it is now easy to show that, for any fixed time t, the quantity $\sum_n P_n(t-s)\langle Z(s)^n \rangle_{\mathrm{LE}}$ is independent of s $(0 \leq s \leq t)$. Evaluating it at $s = t$ and $s = 0$, we find the exact duality relation [47]

$$\int_\ell^1 dz \sum_{n=0}^{\infty} p(z,t) P_n(0) z^n = \int_\ell^1 dz \sum_{n=0}^{\infty} p(z,0) P_n(t) z^n, \tag{8}$$

which also reads $\langle\langle Z(t)^{N(0)}\rangle_{\rm LE}\rangle_{\rm RD} = \langle\langle Z(0)^{N(t)}\rangle_{\rm LE}\rangle_{\rm RD}$, where $\langle\cdot\rangle_{\rm RD}$ has to be understood as an averaging over the RD process. (This relation generalizes an analogous one derived by Doering *et al.* for the reversible coagulation-decoagulation process $A \rightleftharpoons 2A$ [30]).

Using Eq. (8) and properly choosing initial conditions of the LE, one easily computes quantities of the RD process such as the survival probability and the moments of the probability distribution $P_n(t)$. The survival probability $P_{\rm surv}^{(m)}(t)$ is defined as the probability that, starting at $t = 0$ with $m > 0$ particles, at least one particle survives at time t: $P_{\rm surv}^{(m)}(t) = 1 - P_0(t)$. Using Eq. (8) with $p(z, 0) = \delta(z)$ and $P_n(0) = \delta_{mn}$, we obtain

$$1 - P_{\rm surv}^{(m)}(t) = \int_\ell^1 {\rm d}z p(z, t|0, 0) z^m = \langle Z(t)^m\rangle_{\rm LE}, \quad (9)$$

where $p(z, t|z_0, 0)$ is the conditional transition probability of the LE with $p(z, t = 0) = \delta(z - z_0)$ as the initial condition and the mth-order moment is a readily measurable quantity in a LE simulation. Similarly, the moments of the RD process can be derived from $G(z, t)$ using $p(z, 0) = \delta(z - z_0)$ as the initial condition; Eq. (8) yields [48]

$$G(z_0, t) = \int_\ell^1 dz \sum_{n=0}^\infty p(z, t|z_0, 0) P_n(0) z^n. \quad (10)$$

Differentiating $G(z_0, t)$ with respect to z_0 and evaluating it at $z_0 = 1$ yields the (factorial) moments of the RD process. For instance, the average particle number reads

$$\langle N(t)\rangle = \partial_{z_0} \int_\ell^1 dz \sum_n p(z, t|z_0, 0) P_n(0) z^n|_{z_0=1}. \quad (11)$$

All formulas above can be easily generalized to the spatially extended case in the presence of diffusion. For instance, putting m particles at one site i and 0 elsewhere in the RD process and choosing $Z_j(0) = 0$ for all sites j for the LE, Eq. (9) is replaced by $P_{\rm surv}^{(m,i)}(t) = 1 - \langle Z_i(t)^m\rangle_{\rm LE}$ [49].

We now make contact with the field-theoretic approaches alluded to above. In addition to clarifying the situation there, this elucidates the physical meaning of the stochastic variable Z governing Eq. (5) and also reveals how correlation functions at different times and response functions can be calculated within our framework.

We first recall the main features of the Doi-Peliti formalism from which follow the field theories associated with RD processes (see, e.g., [12,15,17,18]). A (state) vector $|P(t)\rangle$ is associated with the set of probabilities $\{P_n(t)\}$. This vector belongs to a Hilbert space spanned by the "occupation number" vectors $\{|n\rangle\}$ and reads $|P(t)\rangle = \sum_{n=0}^\infty P_n(t)|n\rangle$. The vector $|n\rangle$ is an eigenvector with eigenvalue n of the "number" operator $\hat{N} = a^\dagger a$ where a and $a^\dagger$ are annihilation and creation operators satisfying $[a, a^\dagger] = 1$, $a|0\rangle = 0$, $a|n\rangle = n|n-1\rangle$, and $a^\dagger|n\rangle = |n+1\rangle$. The scalar product is chosen such that $\langle m|n\rangle = n!\delta_{mn}$ and $a^\dagger$ is the Hermitian conjugate of a. With any complex number ϕ is associated a coherent state $|\phi\rangle$ defined by the relation $|\phi\rangle = \exp(\phi a^\dagger)|0\rangle$. The probability generating function can then be written as $G(z, t) = \langle z|P(t)\rangle$. The evolution of the $\{P_n(t)\}$ induces the evolution of the state vector, $\partial_t|P(t)\rangle = \mathcal{L}(a^\dagger, a)|P(t)\rangle$, where $\mathcal{L}$, written in its normal-ordered form, is the very same function as in Eq. (2). With the LE probability distribution $p(z, t)$ defined in Eq. (6), we also associate a vector

$$|p(t)\rangle = \int_\ell^1 dz p_c(z, t)|z\rangle + q_1(t)|1\rangle + q_\ell(t)|\ell\rangle, \quad (12)$$

where $|z\rangle$ is the coherent state with real eigenvalue $z \in [\ell, 1]$. From the evolution of $p(z, t)$ one checks that $\partial_t|p(t)\rangle = \mathcal{L}^\dagger(a^\dagger, a)|p(t)\rangle$, where $\mathcal{L}^\dagger(a^\dagger, a)$ is also normal ordered. Using the resolution of the identity $2\pi\mathbb{I} = \int\int_0^\infty dz dz' \exp(-izz')|iz\rangle\langle z'|$, one can show that Eq. (8) reads $\langle p(t)|P(0)\rangle = \langle p(0)|P(t)\rangle$ [32,33]. Thus, within the Doi-Peliti formalism, the duality relation [Eq. (8)] is a direct consequence of the fact that $|P(t)\rangle$ evolves with $\mathcal{L}$ and $|p(t)\rangle$ with $\mathcal{L}^\dagger$.

The field-theoretical approach to RD processes is based on a functional integral representation of the evolution operator $\exp(\mathcal{L}t)$ of the state vector $|P(t)\rangle$. Using the Trotter formula and introducing resolutions of the identity in terms of complex-conjugate coherent states, one can derive the generating functional of correlation and response functions in the presence of real sources [50],

$$\mathcal{Z}[J, \tilde{J}] = \int \mathcal{D}\phi\mathcal{D}\phi^* e^{-S[\phi,\phi^*] + \int dt(J\phi + \tilde{J}\phi^*)}, \quad (13)$$

with the action $S[\phi, \phi^*] = \int dt[\phi^*\partial_t\phi - \mathcal{L}(\phi^*, \phi)]$, where ϕ and ϕ^* are complex-conjugate fields.

The usual derivation of a LE from this field theory goes as follows (see, e.g., [20,25,51]): After performing the shift $\phi^* \to \phi^* + 1$, the fields ϕ and ϕ^* are formally replaced in the action S by a real field ψ, dubbed the density field, and an imaginary field $\tilde{\psi}$, called the response field. For binary reactions, $\mathcal{L}(\tilde{\psi} + 1, \psi)$ is at most quadratic in $\tilde{\psi}$, the term $\exp[\int dt\tilde{\psi}^2 U(\psi)]$ with $U(\psi) = \alpha_2\psi - (\beta_0 + \beta_1)\psi^2$ in $\mathcal{Z}$ is formally written as a Gaussian integral $\int \mathcal{D}\eta \exp(-\int dt[\eta^2/2 + \sqrt{2U(\psi)}\tilde{\psi}\eta])$, and the resulting argument of the exponential is thus linear in $\tilde{\psi}$. The functional integral on the imaginary field $\tilde{\psi}$ then yields

$$\mathcal{Z}[J, \tilde{J}] = \int \mathcal{D}\psi\mathcal{D}\eta\mathcal{P}(\eta(t))\delta(f(\psi, \eta, \tilde{J}))e^{\int dtJ\psi} \quad (14)$$

where $\mathcal{P}(\eta(t)) = \exp(-\int dt\eta(t)^2/2)$, $f = -\partial_t\psi + \alpha_2\psi - (2\beta_0 + \beta_1)\psi^2 + \tilde{J} + \sqrt{2[\alpha_2\psi - (\beta_0 + \beta_1)\psi^2]}\eta$, and $\delta(f(\psi, \eta, \tilde{J}))$ is a functional Dirac function. Written under this form, $\mathcal{Z}[J, \tilde{J}]$ is the generating functional of correlation

functions derived from the LE, $f(\psi,\eta,\tilde{J})=0$, where $\eta(t)$ is interpreted as a Gaussian white noise and the derivation above follows the standard Martin–Siggia–Rose–De Dominicis–Janssen (MSRDJ) method in the reverse order [52–54]. For instance, for pure annihilation ($2A\to\emptyset$), this yields the imaginary noise LE,

$$\partial_t\psi=-2\beta_0\psi^2+i\sqrt{2\beta_0}\psi\eta. \tag{15}$$

The problem with this derivation is that it is purely formal. Although exact to all orders of perturbation theory [34], it is actually incorrect to trade the two complex-conjugate fields ϕ and ϕ^* for a real and an imaginary field in $\mathcal{Z}[J,\tilde{J}]$ because the resulting functional integral is in general no longer convergent at large fields (a fact that is immaterial within perturbation theory). Indeed, the leading term at large fields $-\beta_1\psi^2\tilde{\psi}^2$ has the wrong sign since $\tilde{\psi}$ is purely imaginary, contrary to the original term $-\beta_1\phi^2\phi^{*2}$. The imaginary noise in Eq. (15) is a consequence of this formal and incorrect step. Notice that the derivation is performed assuming that $\psi(t)$ is real, while a field evolving according to Eq. (15) necessarily becomes complex.

To overcome the convergence problems discussed above and the fact that the action S is not quadratic in ϕ^* when reactions $A\to pA$ with $p>2$ are involved, we now provide a proper and general field-theoretical derivation of a LE by using contour deformations in the complex plane. In contrast to the usual procedure, ϕ is deformed into a purely imaginary variable ψ, and the conjugated field ϕ^* into a real variable $\tilde{\psi}\in[\ell,1]$. A complete derivation for a particular RD process is presented in the Supplemental Material [34]. In the spatially extended case, where particles diffuse with rate D, the result reads

$$\mathcal{Z}[J,\tilde{J}]=\int_{\ell}^{1}\mathcal{D}\tilde{\psi}\int_{-i\infty}^{i\infty}\mathcal{D}\psi e^{-S[\psi,\tilde{\psi}]+\int_{t,x}(J\psi+\tilde{J}\tilde{\psi})}, \tag{16}$$

where the functional form of S has not changed (it still involves the same function $\mathcal{L}$ as above, but its arguments are different). Finally, using the same step as discussed above (the MSRDJ formalism in the reverse order), consisting of (i) rewriting the quadratic term in ψ as a functional integral over a Gaussian field η and (ii) integrating over the imaginary variable ψ, we obtain $\mathcal{Z}[J,\tilde{J}]$ under a form similar to Eq. (14) with ψ replaced by $\tilde{\psi}$ and $f(\psi,\eta,\tilde{J})$ replaced by $g(\tilde{\psi},\eta,J)=\partial_t\tilde{\psi}+D\nabla^2\tilde{\psi}+A(\tilde{\psi})+J+\sqrt{2B(\tilde{\psi})}\eta$. Notice that the integration over ψ leading to $\delta(g)$ requires an integration by parts in S of the $\tilde{\psi}\partial_t\psi$ term that changes its sign. A change of t into $-t$ is thus necessary to obtain the usual diffusion term, as can be seen on the explicit expression of g above. Therefore, the corresponding LE for $\tilde{\psi}$ runs backwards in time. Setting $J=0$ and defining $Z(t)\equiv\tilde{\psi}(-t)$, we finally obtain exactly the same LE as Eq. (5).

The derivation above shows that the field $Z(t)$ of the LE (5) is closer in spirit to the (time-reversed) response field than to the direct (density) field, and also that the source term $\int J\psi$ in $\mathcal{Z}[J,\tilde{J}]$—which is convenient to derive correlation functions but may seem unphysical—has a simple meaning here: it appears as an external force in (5), which can then be used to calculate not only response functions but also all correlation functions [55].

As a specific example of the method, we consider the reactions $2A\overset{\beta_0}{\to}\emptyset$ and $A\overset{\alpha_2}{\to}2A$, together with diffusion at rate D. This RD process is archetypical of the prominent directed percolation (DP) universality class [2,3]. Through the Doi-Peliti formalism, the action is readily derived. Using the contour deformations described in [34], and defining $Z(x,t)=\tilde{\psi}(x,-t)$ we obtain

$$\partial_t Z=D\nabla^2 Z-\alpha_2 Z(1-Z)+\sqrt{2\beta_0(1-Z^2)}\eta \tag{17}$$

with $Z=Z(x,t)\in[-1,1]$ (as determined by the contour deformations), which is identical to Eq. (5) for this set of reactions (supplemented with diffusion). For DP, the action is quadratic both in ϕ and ϕ^*, and another contour deformation [58] leads to a LE on the other field $\psi(x,t)$,

$$\partial_t\psi=D\nabla^2\psi+\psi(\alpha_2-2\beta_0\psi)+\sqrt{2\psi(\alpha_2-\beta_0\psi)}\eta, \tag{18}$$

with now $\psi=\psi(x,t)\in[0,\alpha_2/\beta_0]$. Note that Eqs. (17) and (18) are identical up to a simple linear change of variable, something referred to as rapidity symmetry. Note however that Eq. (18) ceases to exist in the pure annihilation limit $\alpha_2\to 0$ ("imaginary noise") [59]. In contrast, Eq. (17) remains well behaved, and should be considered as the correct LE in such a case.

Another well-established universality class of nonequilibrium absorbing phase transitions corresponds to RD processes where the parity of the (local) number of particles is preserved by the reactions [18,60]. Viewing the particles as domain walls between two symmetric absorbing states, the following phenomenological LE with two symmetric absorbing barriers was postulated in [39]:

$$\partial_t\varphi=D\nabla^2\varphi+(a\varphi-b\varphi^3)(1-\varphi^2)+c\sqrt{1-\varphi^2}\eta. \tag{19}$$

With hindsight, this LE turns out to be the exact (time-reversed) response-field LE for the RD process $A\overset{\alpha_3}{\to}3A$, $A\overset{\alpha_5}{\to}5A$, $2A\overset{\beta_0}{\to}\emptyset$, which indeed belongs to this class. Last but not least, it provides us with the microscopic parameter identifications $a=-(\alpha_3+\alpha_5)$, $b=\alpha_5$, and $c=\sqrt{2\beta_0}$.

To summarize, we derived exact LEs for RD processes with at most bimolecular reactants and showed how they can be used to calculate usual quantities of interest. Our work clarifies several misunderstandings that have haunted the related field-theoretical literature for decades, i.e., the

reasons why some RD processes lead to imaginary noise, why some can be written in terms of the so-called density field whereas others can only be written in terms of the response field, and how keeping track of the evolution of the latter allows for computing correlation functions of the original RD process.

Beyond its obvious importance in physics, chemistry, and the many fields where problems can be explicitly formulated in the form of RD processes, our work may have particular impact for the numerous situations where equations similar to Eq. (5) have been written [29,40–43,61–66]. We hope our results will be put into practice and that they will help strengthen and clarify the use of Langevin equations in various fields.

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[49] Because of homogeneity, the mth-order moment at site i can in fact be estimated in a numerical simulation of the LE as an average over all sites: $1/L^d \sum_i < Z_i(t)^m >_{\mathrm{LE}}$.
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