Autocorrelation Functions of Driven Reaction-Diffusion Processes

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We study the effect of bias diffusion on the autocorrelation functions of the one-dimensional annihilation reaction $A + A \to$ inert. Exact results are given for a subset of transition probability rates. Unlike equal-time functions, the interplay between hard-core interactions and the drift velocity gives rise to a rich nonequilibrium behavior of autocorrelation and intermediate scattering functions. Our results are supported and compared with Monte Carlo simulations.

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Reaction-diffusion (RD) processes have been extensively investigated in recent years, giving rise to a vast body of work [1]. In lower dimensions they provide relevant examples of strongly fluctuating systems which are no longer describable by standard mean-field-like chemical rate equations. These models are closely related to coagulation processes [2], random sequential adsorption problems [3], phase separation [4], and Glauber dynamics [5], posing both theoretical and experimental challenges even in $d = 1$. Particular emphasis has been placed on the one-species annihilation process of hard-core particles $A + A \to$ inert. Although many numerical, theoretical [1], and experimental [6] results have been obtained, to the best of our knowledge the analysis of *unequal-time* or autocorrelation functions is still lacking [7]. The understanding of the space and time evolution of these functions plays a particularly important role because its Fourier transform is directly proportional to the scattered intensity in any scattering experiment [8].

As a first step in this direction, we present an exact solution of autocorrelation functions of driven RD processes. The inclusion of a drift velocity would be appropriate for the description of the biased motion of particles in the presence of a force field, a feasible experimental scenario. Using periodic boundary conditions (PBC's), unlike equal-time or instantaneous many-body correlators it will turn out that the presence of bias diffusion induces rather unusual features in the behavior of autocorrelation functions. As in most exact solutions of nonequilibrium dynamics our analysis will be limited to one-dimensional lattices. Nonetheless, such exact solutions become most valuable to elucidate the actual asymptotic dynamics of strongly fluctuation-dominated regimes, providing a reference for more complicated and realistic systems.

The outline of this work is as follows. First, we build up the evolution operator H resulting from to the master equation [9] in terms of the action of a quantum spin "Hamiltonian," since any possible configuration $|s\rangle$ can be expressed in a (pseudo)spin-$\frac{1}{2}$ representation. A particle or vacancy at each of the N-lattice sites corresponds to spin up or down, say. The hard-core character of the A particles prevents double occupancy per site. Second, notice that H is entirely defined by the set of transition probability rates $\{W(s \to s')\}$, which in our driven RD model is given by the following single-step processes. (i) Annihilation of two particles with rate R, lying on a randomly chosen lattice bond, and (ii) right (left) particle hopping (exchange) with probability h (h') within a given bond. Third, H can be cast in terms of a non-Hermitian problem of interacting spinless fermions via a Jordan-Wigner transformation [10]. In order to avoid difficulties with an otherwise singular Bogoliubov [11] similarity transformation it is convenient to introduce a *fictitious* single-step process, namely, (iii) attempts to create A-particle pairs with rate ϵ on a randomly selected bond. This artificial transition rate ϵ will be set to zero at the very end of the calculation. We refer the reader to Ref. [12] for a more detailed derivation. Fourth, we impose the special relationship between transition rates $R + \epsilon = h + h'$ (becoming $R = h + h'$ when the original RD process is recovered under $\epsilon \to 0$). To our knowledge no way of solving the general situation, i.e., arbitrary R, h, and h' is presently known. This constraint removes many fermion terms from the Hamiltonian and allows exact solution [13]. Finally, using PBC's and after Fourier transforming, we map our original evolution operator onto a quadratic form of running-wave fermions which can be diagonalized by a Bogoliubov-type similarity transformation. Thus, finally we are left with the following free fermion "Hamiltonian":

$$H = \sum_{q \in Q} \lambda_q \, \xi_q^+ \, \xi_q \,, \quad \lambda_q = b - a \cos q + i\, v \sin q \,, \tag{1}$$

where $Q = \{\pm\pi/N, \pm 3\pi/N, \ldots, \pm(N-1)\pi/N\}$, $a = R - \epsilon$, $b = R + \epsilon$, and $v = h - h'$ is the drift velocity. Here the ξ operators satisfy standard fermionic anticommutation rules; however, it should be noticed that $\xi_q^+ \neq \xi_q^\dagger$, where $\dagger$ denotes Hermitian conjugation (see Ref. [12]). The limit $\epsilon \to 0$ is rather special in that it gives rise to low-lying gapless modes which are ultimately responsible for the slow asymptotic diffusive dynamics.

We are especially interested in understanding to what extent the nonequilibrium correlations are affected by the

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interplay between hard-core on-site repulsion and the drift velocity v. As is known [9], for a given initial state $|\varphi_0\rangle$, the conditional probability to observe a particle at time $t + t_0$ at site l, given that site m was already occupied by another (or the same) particle at time t_0 (i.e., the autocorrelation function), is given by

$$A_{l,m}(t, t_0) = \langle \tilde{\psi} | \hat{n}_l \, e^{-Ht} \, \hat{n}_m \, e^{-H t_0} \, | \varphi_0 \rangle , \qquad (2)$$

where $\langle \tilde{\psi} |$ is the left steady or left vacuum state of H [14], whereas $\hat{n}_j$ denotes the occupation number operator at site j. Using the Bogoliubov transformation angles

$$\tan 2\theta_q = \frac{2\sqrt{R\,\epsilon}\ \sin q}{b\,\cos q - a} , \qquad (3)$$

it can be shown [12] that in the ξ representation $\hat{n}_j$ can be rewritten as

$$\hat{n}_j = \frac{1}{N} \sum_{k,k' \in Q} e^{i(k'-k)j} (\cos\theta_k \, \xi_k^+ - \sin\theta_k \, \xi_{-k}) \times (\cos\theta_{k'} \, \xi_{k'} - \sin\theta_{k'} \, \xi_{-k'}^+) . \qquad (4)$$

For the sake of simplicity we shall consider an initial state $|\varphi_0\rangle$ corresponding to a full lattice, a common starting point within the context of RD processes; then $\hat{n}_j |\varphi_0\rangle = |\varphi_0\rangle$, $j = 1, 2, \dots, N$. It is a simple matter to check that in terms of the ξ fermions this initial state adopts the form

$$|\varphi_0\rangle = \prod_{q \in P} (1 + \cot\theta_q \ \xi_{-q}^+ \, \xi_q^+) \, |\psi\rangle , \qquad (5)$$

where $|\psi\rangle$ is the right vacuum (steady) state of H [14], and P denotes the set of positive values of $q \in Q$. From Eqs. (1) and (5) it follows that the stochastic evolution of $|\varphi_0\rangle$ at time t_0 then yields

$$e^{-Ht_0} |\varphi_0\rangle = \left[1 + \sum_n \frac{1}{n!} \sum_{q_1 \in P} \cdots \sum_{q_n \in P} \left(\prod_{j=1}^{n} e^{-\gamma_{q_j} t_0} \times \cot\theta_{q_j} \ \xi_{-q_j}^+ \, \xi_{q_j}^+ \right) \right] |\psi\rangle , \quad \gamma_q = 2 \,\mathrm{Re}\, \lambda_q . \qquad (6)$$

This expansion entails immediate consequences for instantaneous ($t = 0$) k-point correlation functions of the form $\langle \tilde{\psi} | \hat{n}_{j_1} \cdots \hat{n}_{j_k} e^{-Ht_0} | \varphi_0 \rangle$. Since the drift velocity enters neither the form (4) nor (6), it is clear that the k-point correlators are *independent* of the bias. Actually, this observation is of rather general character [15]. Hence, for random initial conditions the instantaneous correlations of the system cannot develop any kind of shock wave, though it should be noticed that this situation changes dramatically if *open* boundary conditions are imposed [16]. However, the biased diffusion does affect the time development of autocorrelation functions in a rather involved way, even using PBC's.

For the initially full state $|\varphi_0\rangle$, the calculation of Eq. (2) requires taking into account at most the second order contribution of Eq. (6) as the occupation number operators (4) involve only two ξ fermions. Using the anticommutation rules of these latter operators the actual evaluation of (2) is straightforward albeit rather tedious and lengthy [17]. Taking the limit $\epsilon \to 0$ at the *end* of the calculation, and after introducing the arguments

$$\varphi_n(q) = n\,q - v\,t \sin q , \quad n = l - m \in \mathcal{Z} , \qquad (7)$$

along with the well-defined integrals

$$F_n(t, t_0) = \frac{1}{\pi} \int_0^{\pi} e^{-\gamma_q (t_0 + t/2)} \cos\varphi_n(q) \, dq , \qquad (8)$$

$$G_n(t, t_0) = \frac{1}{\pi} \int_0^{\pi} e^{-\gamma_q (t_0 + t/2)} \tan\frac{q}{2} \ \sin\varphi_n(q) \, dq , \qquad (9)$$

$$H_n(t, t_0) = \frac{1}{\pi} \int_0^{\pi} e^{-\gamma_q (t_0 + t/2)} \cot\frac{q}{2} \ \sin\varphi_n(q) \, dq , \qquad (10)$$

the final result turns out to be

$$A_n(t, t_0) = F_n(t, t_0) [F_n(t, t_0) - F_n(t, 0)] - G_n(t, t_0) \times [H_n(t, t_0) - H_n(t, 0)] + \rho_{t_0} \, \rho_{t+t_0} . \qquad (11)$$

Here $\rho_{t'} = e^{-2Rt'} I_0(2Rt')$ denotes the density at time t', whereas $I_0(z)$ is a modified Bessel function of the first kind. Because of the initial configuration, both density and autocorrelation functions remain translationally invariant $\forall\, t > 0$, as expected. However, notice that $A_n \neq A_{-n}$ if $v \neq 0$. Our results are shown in Figs. 1(a) and 1(b), where we display, respectively, the self-correlation ($n = 0$) and autocorrelation functions.

We have conducted Monte Carlo simulations to test our theoretical expectations in a periodic chain of $N = 10^5$ sites. The minimum attainable time step measurement is therefore $1/N$. Using the microscopic single-step rules described above (with $\epsilon \equiv 0$), we obtained an excellent agreement with Eq. (11) by averaging over 10^3 independent histories of an initially full lattice (see Fig. 1). Similar numerical results were obtained starting from an initially *random* distribution with density ρ_{t_0}. For $v \neq 0$ the self-correlations exhibit a nonmonotonic *crossover* from an exponential regime to a diffusive dominated asymptotic kinetics. More specifically, for $R\,t_0 \gg 1$, $A_0(t, t_0)$ behave as

$$A_0(t, t_0) \simeq \begin{cases} \rho_{t_0} e^{-Rt} + O(t^2) , & R\,t \ll 1 , \\ \rho_{t_0} / \sqrt{4\pi R t} + O(t_0 / t^{3/2}) , & t \gg t_0 . \end{cases} \qquad (12)$$

The self-correlations of the isotropic RD system ($v = 0$) do not show such a crossover, and decay diffusively in a monotonic form.

To understand the space and time development of autocorrelations ($n \neq 0$), we have carried out an asymptotic expansion of the integrals (8), (9), and (10), in which the small q regime of integration dominates and which is valid if any one (or more) of $R\,t$, $R\,t_0$, or n is large. It turns out that then $G \ll F \sim H$ so the middle term in Eq. (11) is negligible, and

$$F_n(t, t_0) \simeq \frac{e^{-(n-v\,t)^2/4R\,\tau}}{2\sqrt{\pi R \tau}}, \tag{13}$$

where $\tau \equiv t_0 + t/2$. The "ballistic" $n - v\,t$ factor arises from $\varphi_n(q)$ [Eq. (7)] and the form of (13), which is analogous to wave packet spreading for a free particle in quantum mechanics. Equation (13) explains for large n the peak seen for positive n in Fig. 1(b) at $t = n/v$ and its width $\frac{1}{v}\sqrt{R\,(t_0 + n/v)}$ (in time t). The smooth background in Fig. 1(b) is from the $\rho_{t_0}\,\rho_{t+t_0}$ term in Eq. (11). Also, (13) yields for $Rt_0 \gg 1$ the time t^* at which the dip occurs in Fig. 1(a), namely, $t^* \sim \frac{3R}{v^2}\ln(t_0/t^*)$. Notice that *connected* autocorrelations, i.e., $C_n(t, t_0) = A_n(t, t_0) - \rho_t\,\rho_{t+t_0}$, decay asymptotically as $C_n(t, t_0) \simeq t_0/(2\pi R\,t^2)\,e^{-(n-v\,t)^2/R\,t}$ [7].

Rather than considering the autocorrelation functions in real space, it is often more convenient to focus attention on the Fourier transform components of the connected autocorrelations, namely, $S_0(q, t) = \sum_{n \in Z} C_n(t, t_0)\,e^{iqn}$. These are usually called intermediate scattering functions (ISF) [8] and are closely related to measurements of light scattering in *real* RD systems [6]. Our analysis suggests that sufficiently diluted RD systems ($Rt_0 \gg 1$) could exhibit modulated liquidlike structures within nonequilibrium regimes, particularly at early evolution stages. This is illustrated in Fig. 2. The *phase* periodicity of ISF functions decreases with the bias in a rather complex form as is shown in Figs. 2(a) and 2(b). However, the *amplitudes* of ISF are independent of the bias $\forall\ t$, t_0, q [see Fig. 2(c)].

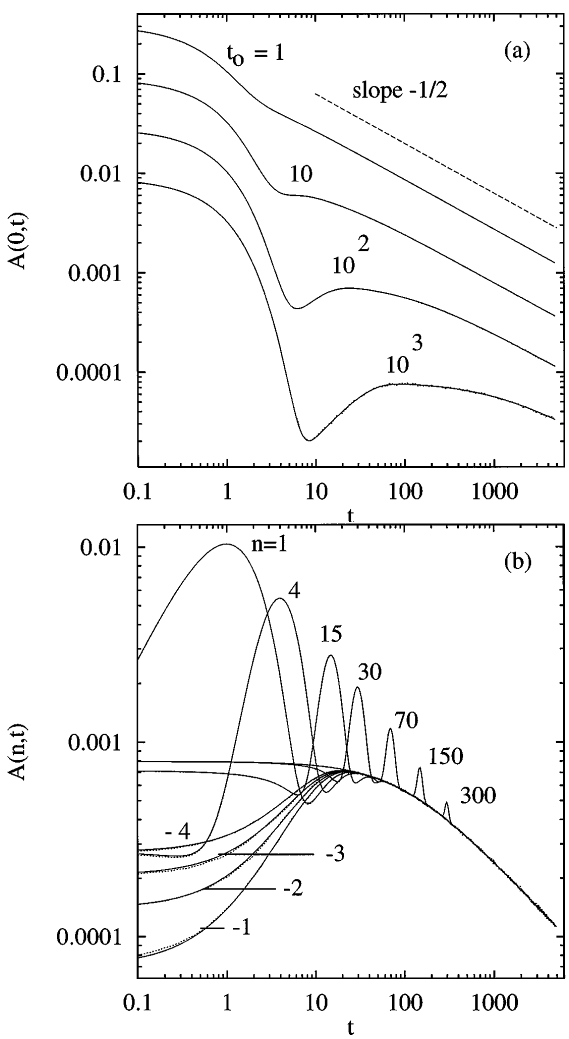

FIG. 1. Autocorrelation functions starting from an initially full lattice. Solid lines refer to Eq. (11) in the text using $R = h = 1$. Dotted lines (slightly observable) denote our numerical results averaged over 10^3 Monte Carlo runs, for a periodic lattice of 10^5 sites. (a) Self-correlation functions for different initial relaxation times. (b) Results for autocorrelation functions using $t_0 = 10^2$.

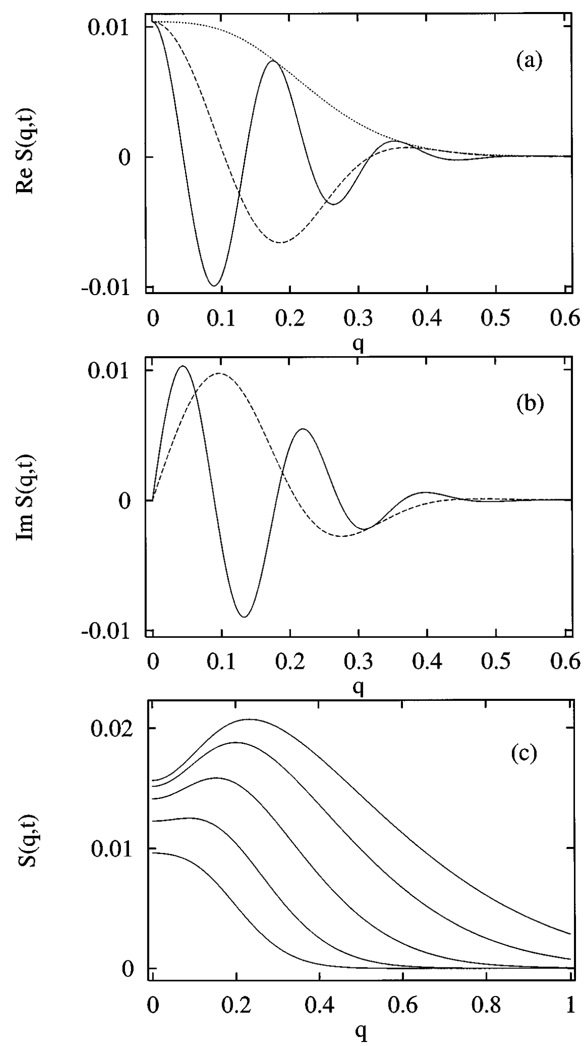

FIG. 2. Intermediate scattering functions for $R = 1$, $t_0 = 10^2$, and $t = 50$. (a) Real and (b) imaginary parts for $v = 0.3$ (dashed lines) and $v = 0.7$ (solid lines). The dotted lines in (a) denote the undriven case. (c) Amplitudes of ISF for $t_0 = 10^2$ and $t = 5, 8, 15, 30,$ and 60 (from top to bottom). The amplitudes are drift independent.

Because of the lattice inversion symmetry, it is clear that $A_n(v) = A_{-n}(-v)$, $\forall\ t,\ t_0$, and therefore, $S(v) = S^*(-v)$. What is more interesting here is the observation that $|S_0(q,t)|$ is preserved regardless the actual current present in the system.

In summary, we have constructed an exact solution for autocorrelation functions of a biased reaction diffusion process, for a particular relationship of transition rates. Monte Carlo simulations confirm the analytic work, and suggest that the results are not substantially changed on relaxing the constraint (see [12] and [13]). A general argument has been given which shows that instantaneous correlations are unaffected by the bias. On the other hand, the bias strongly affects the time-developed autocorrelation functions, leading to propagating structures and other interesting modulations in nonequilibrium regimes. In a recent work [18], it has been shown that different stochastic systems can be treated on an equal footing by means of similarity transformations. Therefore our results could be extended to other equally interesting stochastic processes. Finally, we have also calculated intermediate scattering functions, which capture modulated structures, in the hope that this will stimulate experimental work which could be compared to this theoretical study.

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