Exact time-dependent correlation functions for the symmetric exclusion process with open boundary

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As a simple model for single-file diffusion of hard core particles we investigate the one-dimensional symmetric exclusion process. We consider an open semi-infinite system where one end is coupled to an external reservoir of constant density ρ^* and which initially is in a nonequilibrium state with bulk density ρ_0. We calculate the exact time-dependent two-point density correlation function $C_{k,l}(t) \equiv \langle n_k(t) n_l(t) \rangle - \langle n_k(t) \rangle \langle n_l(t) \rangle$ and the mean and variance of the integrated average net flux of particles $N(t) - N(0)$ that have entered (or left) the system up to time t. We find that the boundary region of the semi-infinite *relaxing* system is in a state similar to the bulk state of a finite *stationary* system driven by a boundary gradient. The symmetric exclusion model provides a rare example where such behavior can be proved rigorously on the level of equal-time two-point correlation functions. Some implications for the relaxational dynamics of entangled polymers and for single-file diffusion in colloidal systems are discussed.

I. INTRODUCTION

The simple symmetric exclusion process (SEP) [1] is a Markov process describing the diffusive motion of identical hard-core particles on a lattice. Particles hop randomly to nearest-neighbor sites with an exponential waiting-time distribution with mean τ_0, provided the chosen site is empty. If it is occupied, the attempt to move is rejected. A considerable body of exact results, particularly for the one-dimensional case (see [2,3] for reviews), has led to a thorough understanding of many fundamental properties of this model. Applications to specific problems include interface growth in the universality class of the Edwards-Wilkinson equation [4,5], reptation dynamics of entangled polymers [6], or single-file diffusion in molecular-sized channels such as biological membrane channels [7] or zeolite pores [8], and in one-dimensional colloidal systems [9].

From a theoretical and also experimental point of view the main quantities of interest are (a) the equilibrium and stationary nonequilibrium properties of the system with open boundaries where particles can enter and leave [10] and (b) the time evolution of the local particle density and their correlations in a system that relaxes after it has been prepared in some far-from-equilibrium initial state. For instance, in the study of kinetic roughening of an initially flat interface in a two-phase Ising system one obtains from the SEP exact universal scaling functions for the roughness [11]. In the investigation of the reptation dynamics [12,13] of an initially stretched DNA chain, flourescence microscopy allows for a direct measurement of the relaxation of the tube length [14] which in the lattice gas approach [15] is proportional to the number of particles in the one-dimensional exclusion process with open boundary. Not only universal power laws, but also nonuniversal amplitudes are of interest, e.g., for estimating the sensitivity of the coarse-graining involved in the lattice gas description to the microscopic properties of the polymer dynamics in a dense solution [6,16].

For a more detailed understanding of the role of correlations in nonequilibrium states of the symmetric exclusion process [25], we compute here the time-dependent two-point density correlation function in an one-dimensional semi-infinite system with one open boundary, connected to a reservoir of constant density ρ^*. In the polymer context this is equivalent to an entropic tensile force acting at the end segments of an entangled polymer chain [13].

The paper is organized as follows. In order to overcome some of the technical difficulties connected with the coupling to boundary reservoirs we formulate the SEP in terms of the dynamical matrix product ansatz [17–19] (Sec. II). In Sec. III we derive a functional equation for the two-point density correlation function that is solved by a Bethe wave function. From this we derive in Sec. IV the scaling form in terms of error functions. The details of these rather involved calculations are presented in the appendixes. In Sec. V we focus on the fluctuations in the total number of particles $Q(t) = N(t) - N(0)$ that enter and leave the system that initially is prepared in an uncorrelated random state with a density ρ_0. We obtain the (expected) universal asymptotic growth law $\langle Q^2 \rangle - \langle Q \rangle^2 = A \sqrt{t/\tau_0}$ and the nonuniversal amplitude $A(\rho_0, \rho^*)$ relevant to reptation dynamics. We conclude with some brief remarks on the nature of the evolving nonequilibrium state (Sec. VI).

II. DYNAMICAL MATRIX PRODUCT ANSATZ

We first consider the symmetric exclusion process on a chain of L sites. At the boundary sites $k = 1, L$ particles are injected (extracted) with rates $\alpha_{1,L}$ ($\gamma_{1,L}$). In terms of the transition rates $w_{n \to n'}$ from state n to n' the stochastic dynamics is described by a master equation

$$\frac{d}{dt} P(\underline{n}; t) = \sum_{\underline{n}' \in X} [w_{\underline{n}' \to \underline{n}} P(\underline{n}'; t) - w_{\underline{n} \to \underline{n}'} P(\underline{n}; t)] \quad (1)$$

for the probability $P(n;t)$ of finding, at time t, a configuration n of particles on a lattice of L sites. Here $n = \{n_1, n_2, \ldots, n_L\}$, where $n_i = 0,1$ are the integer-valued particle occupation numbers at site i. In what follows it is convenient to set the microscopic time unit $\tau_0 = 1$. In these units the single-particle diffusion coefficient is given by $D_0 = 1/2$.

Inserting the transition rates as described above, one readily obtains the equations of motion for m-point joint probabilities $\langle n_{k_1}, \ldots, n_{k_m} \rangle$. These form a hierarchy of coupled equations where the time derivatives of the m-point joint probabilities are coupled to $(m-1)$-point joint probabilities. In principle, this allows for a recursive solution, which, however, is difficult to obtain for $m > 1$. More important, unlike in a periodic system, the lack of translational invariance makes it difficult to obtain exact results even for $m = 2$ that we study in this paper. Hence we reformulate the dynamics in terms of the dynamical matrix product ansatz [17–19]. This ansatz leads to a decoupling of the joint probabilities and splits the many-body dynamical problem into a (trivial) dynamical single-particle part and a (nontrivial) many-body part that is time-independent and can be solved using the Bethe ansatz.

The dynamical matrix product ansatz is reviewed in detail in [3]. Here we discuss only the main features relevant to the SEP. One represents an occupied (vacant) site by a time-dependent matrix D (E) in a string $DDDEDEE\ldots$ of L such matrices. The configurational probabilities $P(n;t)$ are obtained by sandwiching the product of these L matrices D or E between suitably chosen vectors $\langle\langle W|$ and $|V\rangle\rangle$ and normalizing by $Z_L = \langle\langle W|C^L|V\rangle\rangle$, where $C = D + E$. [Notice that expanding the Lth power of C automatically gives the sum of all un-normalized configurational probabilities and hence yields the correct normalization factor.] The time-dependent matrices satisfy algebraic relations that are determined by requiring the matrix product state to satisfy the master equation (1). Expectation values of local observables are obtained by sandwiching suitable products of the matrices D, E with C. Defining formally $D_k = C^{k-1}DC^{-k}$ one obtains for the local particle density $\rho_k(t) = \langle n_k(t) \rangle$ at site k,

$$\rho_k(t) \equiv \langle\langle W|C^{k-1}DC^{L-k}|V\rangle\rangle/Z_L \tag{2}$$

$$= \langle\langle W|D_k C^L|V\rangle\rangle/Z_L \tag{3}$$

and for the joint probability $G_{k,l}(t) = \langle n_k(t)n_l(t) \rangle$ of finding particles at sites k,l,

$$G_{k,l}(t) = \langle\langle W|D_k D_l C^L|V\rangle\rangle/Z_L. \tag{4}$$

From these quantities one obtains the two-point density correlation function

$$C_{k,l}(t) = \langle n_k(t)n_l(t) \rangle - \langle n_k(t) \rangle \langle n_l(t) \rangle. \tag{5}$$

Higher-order joint probabilities are obtained analogously. The initial probability distribution is encoded in the matrices $D(0)$.

We do not review here how the matrix relations and the corresponding relations for the vectors $\langle\langle W|, |V\rangle\rangle$ are obtained from the master equation, but refer the reader to [3]. The matrix C as well as the vectors $\langle\langle W|$ and $|V\rangle\rangle$ may be chosen to be time-independent [19]. The dynamical problem is then solved by introducing Fourier transforms

$$\mathcal{D}_p(t) = \sum_k e^{ipk} D_k(t). \tag{6}$$

They have the simple time-dependence

$$\mathcal{D}_p(t) = e^{-\epsilon_p t} \mathcal{D}_p(0) \tag{7}$$

in terms of the initial matrix $\mathcal{D}_p(0)$ and the inverse relaxation times $\epsilon_p = 1 - \cos p$. These diffusive single-particle relaxation modes reflect the random walk nature of the dynamics. For calculating the local expectation values (3) and (4) it is useful to separate the static ($p = 0$) and dynamical ($p \neq 0$) parts in the Fourier expansion of D_k and to write the inverse Fourier transform in the form

$$D_k(t) = (1-k)\mathcal{D}_0 + \mathcal{I} + \int' \frac{dp}{2\pi} \mathcal{D}_p(0) e^{-ipk - \epsilon_p t}. \tag{8}$$

In the semi-infinite system that we will be considering, the primed integral is to be understood as a contour integral in the variable $z = e^{-ip}$, where the contour is chosen in a way such that the values of the joint probabilities obey the correct initial conditions at $t = 0$. The matrices $\mathcal{D}_0$ and $\mathcal{I}$ are time-independent and yield all stationary expectation values. The time-dependent integral contains the relaxational part of D_k.

The master equation not only determines the time evolution of $\mathcal{D}_p(t)$, which is given by Eq. (7), but also requires the Fourier components of $D_k(t)$, as given by Eq. (8), to satisfy various relations among themselves and with the vectors $\langle\langle W|, |V\rangle\rangle$. The dynamical part, where both p_1 and p_2 are nonzero, satisfies

$$\mathcal{D}_{p_1}\mathcal{D}_{p_2} = S(p_1, p_2)\mathcal{D}_{p_2}\mathcal{D}_{p_1} \tag{9}$$

with

$$S(p_1, p_2) = -\frac{1 + e^{ip_1 + ip_2} - 2e^{ip_2}}{1 + e^{ip_1 + ip_2} - 2e^{ip_1}}. \tag{10}$$

The relations involving static components read

$$[\mathcal{D}_p, \mathcal{D}_0] = 0, \tag{11}$$

$$[\mathcal{D}_p, \mathcal{I}] = 2\mathcal{D}_0 \mathcal{D}_p, \tag{12}$$

$$[\mathcal{D}_0, \mathcal{I}] = \mathcal{D}_0^2. \tag{13}$$

These relations have their origin in the bulk exclusion interaction between particles.

The boundary conditions determine the action of $\mathcal{D}_p$ on the vectors $\langle\langle W|$ and $|V\rangle\rangle$. One finds

$$0 = \langle\langle W|\{\mathcal{D}_0 + 2(\alpha_1 + \gamma_1)\mathcal{I} - 2\alpha_1\}, \tag{14}$$

$$0 = \langle\langle W|\{\mathcal{D}_p + e^{2ip}B_1(p)\mathcal{D}_{-p}\} \quad (p \neq 0), \tag{15}$$

$$0=\{(2\alpha_L+2\gamma_L-1)\mathcal{D}_0+2(\alpha_L+\gamma_L)\mathcal{I}-2\alpha_L\}|V\rangle\rangle, \tag{16}$$

$$0=\{B_L(p)\mathcal{D}_p+\mathcal{D}_{-p}\}|V\rangle\rangle \quad (p\neq0). \tag{17}$$

with

$$B_i(p)=\frac{2\alpha_i+2\gamma_i-1+e^{-ip}}{2\alpha_i+2\gamma_i-1+e^{ip}}. \tag{18}$$

Notice that in the relations (9)–(17) the time-dependence drops out. The set of relations (7), (9)–(18) provides an alternative mathematical formulation of the symmetric exclusion process with open boundaries.

III. CORRELATION FUNCTIONS

For the equilibrium choice of boundary parameters $\alpha\equiv\alpha_1=\alpha_L=\rho^*/2$ and $\gamma\equiv\gamma_1=\gamma_L=(1-\rho^*)/2$ the boundary relations (14)–(17) and the functions $B_i(p)$ simplify considerably. With these rates, modeling the connection to particle reservoirs of density ρ^*, the (unique) invariant measure of the process is a product measure with density ρ^*, i.e., there are no density correlations between different sites. Choosing as initial state an uncorrelated state with density $\rho_0\neq\rho^*$ leads to a nontrivial time evolution as the system starts to fill up ($\rho_0<\rho^*$) or deplete (respectively $\rho_0>\rho^*$). Correlations are built up in the transient regime before the equilibrium state is attained. In a semi-infinite system this will take an infinite amount of time, and the "transient" regime is the only relevant one. For finite systems with L sites [10] the system is transient for times $t<\tau^*\propto L^2$. This is the regime on which we focus our attention. Therefore we ignore the right boundary site by considering the thermodynamic limit $L\to\infty$. The only remaining length scale (besides the unit lattice constant) is then the dimensionless diffusion length

$$\tilde{L}\equiv\tilde{L}(t)=\sqrt{4t/\pi\tau_0}. \tag{19}$$

This is a dynamical length scale playing the role of a correlation length (see below).

Anticipating the importance of macroscopic *static* initial and equilibrium properties for the nonequilibrium relaxation process we introduce the basic quantities characterizing both the nonequilibrium initial state with density ρ_0 and the asymptotic equilibrium state with density ρ^*. These are the density gradient

$$\Delta\rho=\rho^*-\rho_0 \tag{20}$$

between bulk and boundary and the compressibility

$$\kappa=\lim_{L\to\infty}(\langle N^2\rangle-\langle N\rangle^2)/L, \tag{21}$$

which is readily obtained from the static two-point correlation function

$$C_{k,l}=\rho(1-\rho)\delta_{k,l} \tag{22}$$

of the uncorrelated initial and final distributions, respectively. Here $\delta_{k,l}$ is the Kronecker delta-function. Hence

$$\kappa=\rho(1-\rho), \tag{23}$$

where $\rho=\rho_0$ or ρ^*, respectively.

A. Density profile and current

The evolving density profile was computed exactly in Ref. [17]. If $\alpha_1=\alpha_L=\rho^*/2$, $\gamma_1=\gamma_L=(1-\rho^*)/2$, one has

$$\rho_k(t)=\rho^*+\int'\frac{dp}{2\pi}\langle\langle\mathcal{D}_p\rangle\rangle e^{-ipk-\epsilon_p t}, \tag{24}$$

where we use the abbreviation

$$\langle\langle\mathcal{D}_{p_1},\ldots,\mathcal{D}_{p_N}\rangle\rangle\equiv\frac{\langle\langle W|\mathcal{D}_{p_1},\ldots,\mathcal{D}_{p_N}C^L|V\rangle\rangle}{Z_L} \tag{25}$$

in order to make the formulas more compact. Using Eqs. (15) and (17) with $B_1(p)=B_L(p)=e^{-2ip}$, one can show that $\langle\langle\mathcal{D}_p\rangle\rangle$ obeys the functional equation $\langle\langle\mathcal{D}_p\rangle\rangle=-\langle\langle\mathcal{D}_{-p}\rangle\rangle$, with the solution

$$\langle\langle\mathcal{D}_p\rangle\rangle=\sum_{k_0=1}^{L}a_{k_0}(e^{ipk_0}-e^{-ipk_0}), \tag{26}$$

where the constants a_{k_0} are determined by the initial conditions. Furthermore, Eqs. (15) and (17) impose constraints on the set of allowed momenta, given by $e^{2ip(L+1)}=1$. In the thermodynamic limit $L\to\infty$, the momenta p form a continuous set and this condition can be relaxed. For a system that is initially in an uncorrelated state with density ρ_0, one has $a_{k_0}=-\Delta\rho$ and one obtains in the thermodynamic limit, after substitution of Eq. (26) in Eq. (24),

$$\rho_k(t)=\rho_0+(\rho^*-\rho_0)g_k(t), \tag{27}$$

where $g_k(t)$ is the lattice analog of the complementary error function

$$g_k(t)=e^{-t}\left[I_k(t)+2\sum_{p=k+1}^{\infty}I_p(t)\right] \tag{28}$$

and where $I_k(t)$ are the modified Bessel functions (A1). In terms of the scaling variable

$$\tilde{x}=k/\tilde{L} \tag{29}$$

the long-time behavior of the density profile is given by the error function, as is well-known for diffusive transport. In the vicinity of the boundary, i.e., at distances $\tilde{x}\ll1$ small compared to the diffusion length, the density profile is linear.

Associated with the spatial variation of the density there is a diffusive relaxational current $j_k=D_0(\rho_k(t)-\rho_{k+1}(t))$ that is space-independent close to the boundary to lowest leading order in time. It is convenient to define the current

$$\hat{j}=j/D_0 \tag{30}$$

in units of the single-particle diffusion coefficient $D_0 = 1/(2\tau_0)$. From the expansion (A2) of the Bessel function one finds

$$\hat{j} = 2(\rho^* - \rho_0)/\sqrt{2\pi t} = 2\sqrt{2}\Delta\rho/(\pi\tilde{L}). \qquad (31)$$

B. Two-point correlation function: Exact expression

The density profile $\rho_k(t)$ and hence the time-dependent *equilibrium* two-point correlation functions $C_{k,l}^*(t) = \langle n_k(t)n_l(0)\rangle - (\rho^*)^2$ can be obtained in a straightforward manner from the solution of a lattice diffusion equation. The solution of the equations of motion for equal-time joint probabilities, however, and hence the calculation of the time-dependent two-point correlation function (5) is much more involved. A convenient way to circumvent an explicit integration of the coupled equations is to make use of the algebraic representation of expectation values within the dynamical matrix product ansatz. Substituting Eq. (8) in Eq. (4) and using the commutation relations (11)–(13), one obtains, when $\alpha_1 = \alpha_L = \rho^*/2$, $\gamma_1 = \gamma_L = (1-\rho^*)/2$,

$$\langle n_k(t)n_l(t)\rangle = \rho^{*2} + \rho^* \int' \frac{dp}{2\pi}\langle\langle \mathcal{D}_p\rangle\rangle e^{-ipk-\epsilon_p t}$$

$$+ \rho^* \int' \frac{dp}{2\pi}\langle\langle \mathcal{D}_p\rangle\rangle e^{-ipl-\epsilon_p t}$$

$$+ \int' \int' \frac{dp_1}{2\pi}\frac{dp_2}{2\pi}\langle\langle \mathcal{D}_{p_1}\mathcal{D}_{p_2}\rangle\rangle$$

$$\times \exp[-i(p_1 k + p_2 l) - (\epsilon_{p_1} + \epsilon_{p_2})t], \qquad (32)$$

where $\langle\langle \mathcal{D}_p\rangle\rangle$ is given by Eq. (26).

Using Eqs. (9) and (15), we can show that $\langle\langle \mathcal{D}_{p_1}\mathcal{D}_{p_2}\rangle\rangle$ obeys the following functional equation

$$\langle\langle \mathcal{D}_{p_1}\mathcal{D}_{p_2}\rangle\rangle = -S(-p_1,p_2)\langle\langle \mathcal{D}_{p_2}\mathcal{D}_{-p_1}\rangle\rangle \qquad (33)$$

together with similar equations involving all the possible arrangements of p_1 with p_2 or $-p_2$ and $-p_1$ with p_2 or $-p_2$. Furthermore, in a finite system, these equations and equation (17) determine the set of allowed momenta. These equations are the equations obeyed by the Bethe wave function of a quantum spin 1/2 system with boundary fields [20] and they have the solution

$$\langle\langle \mathcal{D}_{p_1}\mathcal{D}_{p_2}\rangle\rangle = \sum_{k_0 < l_0}^{L} a_{k_0,l_0}\Psi_{p_1,p_2}(k_0,l_0), \qquad (34)$$

where the constants a_{k_0,l_0} are determined by the initial conditions and $\Psi_{p_1,p_2}(k_0,l_0)$ is the Bethe wave function, which is given by

$$\Psi_{p_1,p_2}(k_0,l_0) = \exp(ip_1 k_0 + ip_2 l_0) + S(p_1,p_2)\exp(ip_2 k_0$$

$$+ ip_1 l_0) - \exp(-ip_1 k_0 + ip_2 l_0)$$

$$- S(-p_1,p_2)\exp(ip_2 k_0 - ip_1 l_0)$$

$$- S(p_1,p_2)\exp(-ip_2 k_0 + ip_1 l_0)$$

$$+ S(-p_1,p_2)\exp(-ip_2 k_0 - ip_1 l_0)$$

$$- S(-p_1,p_2)S(p_1,p_2)\exp(ip_1 k_0 - ip_2 l_0)$$

$$+ S(-p_1,p_2)S(p_1,p_2)$$

$$\times \exp(-ip_1 k_0 - ip_2 l_0). \qquad (35)$$

Using Eq. (24), one can write the two-point correlation function as

$$C_{k,l}(t) = \int' \int' \frac{dp_1}{2\pi}\frac{dp_2}{2\pi}(\langle\langle \mathcal{D}_{p_1}\mathcal{D}_{p_2}\rangle\rangle - \langle\langle \mathcal{D}_{p_1}\rangle\rangle$$

$$\times \langle\langle \mathcal{D}_{p_2}\rangle\rangle)\exp[-i(p_1 k + p_2 l) - (\epsilon_{p_1} + \epsilon_{p_2})t], \qquad (36)$$

where $\langle\langle \mathcal{D}_{p_1}\mathcal{D}_{p_2}\rangle\rangle$ is given by (34) and $\langle\langle \mathcal{D}_{p_1}\rangle\rangle$ by Eq. (26).

If we now choose the contour of integration such that at $t=0$ the integral over $\Psi_{p_1,p_2}(k_0,l_0)$ in Eq. (36) is equal to $\delta_{k,k_0}\delta_{l,l_0}$, then the condition that the initial state is uncorrelated $C_{k,l}(0) = 0$, gives $a_{k_0,l_0} = a_{k_0}a_{l_0} = (\Delta\rho)^2$. Substituting this result in Eq. (34) and using this equation, together with Eq. (26) in Eq. (36), we obtain for the semi-infinite system, when $L \to \infty$,

$$C_{k,l}(t) = (\Delta\rho)^2 \Bigg\{ e^{-2t}[I_{l+k}(2t) - I_{l-k}(2t)]$$

$$+ \sum_{k_0 < l_0}^{\infty} \int' \int' \frac{dp_1}{2\pi}\frac{dp_2}{2\pi}\exp[-i(p_1 k + p_2 l)$$

$$- (\epsilon_{p_1} + \epsilon_{p_2})t][\{S(p_1,p_2) - 1\}\exp(ip_2 k_0 + ip_1 l_0)$$

$$- \{S(-p_1,p_2) - 1\}\exp(ip_2 k_0 - ip_1 l_0) - \{S(p_1,p_2)$$

$$- 1\}\exp(-ip_2 k_0 + ip_1 l_0) + \{S(-p_1,p_2) - 1\}$$

$$\times \exp(-ip_2 k_0 - ip_1 l_0) - \{S(-p_1,p_2)$$

$$\times S(p_1,p_2) - 1\}\exp(ip_1 k_0 - ip_2 l_0) + \{S(-p_1,p_2)$$

$$\times S(p_1,p_2) - 1\}\exp(-ip_1 k_0 - ip_2 l_0)] \Bigg\}, \qquad (37)$$

where the first term comes from the summation terms of $\langle\langle \mathcal{D}_{p_1}\rangle\rangle\langle\langle \mathcal{D}_{p_2}\rangle\rangle$ with $k_0 = l_0$. We remark that for noninteracting particles (no hard-core repulsion and hence no exclusion, but otherwise identical hopping dynamics) the correlation function has only this term, but with a different amplitude $a_0 = \langle n^2\rangle_0 - \rho_0^2 - \rho_0$ determined by the initial distribution only. The exclusion interaction gives rise to the double sum over k_0, l_0 with the terms of the form $S-1$, $SS'-1$.

To extract more detailed information from the exact expression (37), one needs to perform the double sum over k_0, l_0 and to determine the contour of integration such that Eq. (37) obeys the initial condition. This is shown in Appendix B. As a result, we obtain the more compact expression

$$C_{k,l}(t) = -(\Delta\rho)^2 [F_{k+l-1,l-k}(t) + F_{k+l,l-k+1}(t)$$
$$- F_{l-k-1,k+l}(t) - F_{l-k,k+l+1}(t)] \qquad (38)$$

with

$$F_{m,n}(x) = \frac{4 e^{-2x}}{\pi} \int_0^{\pi/2} d\theta \cos\theta \int_0^x dv \, e^{2v\cos^2\theta}$$
$$\times I_m(2(x-v)\cos\theta)\cos[v\sin(2\theta) + n\theta], \qquad (39)$$

which will be used in the next section to obtain the scaling behavior of the two-point correlation function and in Sec. V to determine the particle number fluctuations.

IV. SCALING BEHAVIOR OF THE TWO-POINT CORRELATION FUNCTION

The expression (38) is our starting point for analyzing the particle number fluctuations (see next section). For investigating local correlations it is more convenient to write Eq. (38) in another way

$$C_{k,l}(t) = -(\Delta\rho)^2 [\hat{F}_{k,l-1}(t) + \hat{F}_{k,l}(t) - \hat{F}_{-k,l-1}(t)$$
$$- \hat{F}_{-k,l}(t)] \qquad (40)$$

with

$$\hat{F}_{m,n}(x) = 2t \int_0^1 dv \, e^{-v^2 t} e^{-2t(1-v)}$$
$$\times (1-v)^{n-m+1} I_m(t(1-v)) I_n(t(1-v)). \qquad (41)$$

One can show that Eq. (40) holds if we use the integral representation (A1) of the modified Bessel functions appearing in the definition of $F_{m,n}(x)$, as given by Eq. (39). We can then perform the integral over v. Representing the integral over θ as a complex integral over an appropriate contour we then obtain, after performing some expansions and using the identities (A14) and (A8), $F_{m,n}(x) = \hat{F}_{(m-n+1)/2,(m+n-1)/2}(x)$ which, when substituted in Eq. (38), yields Eq. (40).

For $t \gg 1$ the main contribution to the integral comes from small values of v. In order to obtain the scaling behavior we define an analogy to Eq. (29) a second scaling variable $\tilde{y} = l/\tilde{L}$ and substitute the integration variable $u = v\tilde{L}$. To leading order in time one then has $(1-v)^{k-j} = \exp[-u(\tilde{x} - \tilde{y})]$. With Eq. (A2) we obtain the scaling form of the correlation function
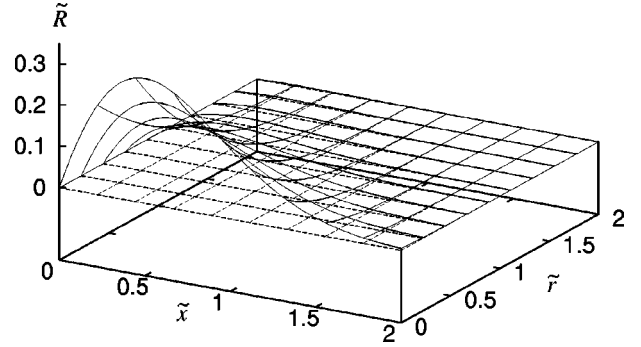

FIG. 1. Scaling part $\tilde{R} = R(\tilde{x}, \tilde{y}) - R(-\tilde{x}, \tilde{y})$ of the two-point correlation function as a function of the scaled bulk coordinate $\tilde{x}$ and the scaled lattice distance $\tilde{r} = \tilde{y} - \tilde{x}$ (full lines). For visualization purposes also the plane $\tilde{R} = 0$ (equilibrium case) is shown (broken lines).

$$C_{\tilde{x},\tilde{y}}(t) = -(\Delta\rho)^2 \sqrt{\frac{\pi}{t}} [R(\tilde{x}, \tilde{y}) - R(-\tilde{x}, \tilde{y})], \qquad (42)$$

where

$$R(\tilde{x}, \tilde{y}) = e^{-(\tilde{x}+\tilde{y})^2/\pi} \operatorname{erfc}((\tilde{y} - \tilde{x})/\sqrt{\pi}) \qquad (43)$$

and $\operatorname{erfc}(\cdot)$ is the complementary error function (Fig. 1). As time increases, the range of correlations increases proportionally to $\sqrt{t}$, but the amplitude decreases in the same manner. The negative sign of the correlation function signals anticorrelations typical of the exclusion effect [2]. In a finite system with a fixed density gradient between the two boundaries imposed by the coupling to two different particle reservoirs these anticorrelations persist [10]. They extend over the whole lattice and have an amplitude inversely proportional to the system size.

In the vicinity of the boundary ($0 \leq \tilde{x} < \tilde{y} \ll 1$) one has $R(\tilde{x}, \tilde{y}) - R(-\tilde{x}, \tilde{y}) = 4\tilde{x}(1-\tilde{y})/\pi$. In terms of the current (31) and the diffusion length $\tilde{L}(t)$ (19) the boundary correlation function is thus given by

$$C = -\tilde{L}\hat{j}^2 \tilde{x}(1 - \tilde{y}). \qquad (44)$$

Corrections, which can easily be obtained from the exact scaling function (42), are of third order in the scaling variables.

In order to disentangle the effects caused by the initial distribution and the exclusion interaction, a comparison with noninteracting particles is again instructive. The scaling form of the correlation function is similar in structure, but one has $R(\tilde{x}, \tilde{y}) = e^{-(\tilde{x}-\tilde{y})^2/\pi}$ and a different amplitude $a_0/\sqrt{4\pi t}$ [see remark after Eq. (37)]. For the same uncorrelated initial distribution as considered for the exclusion process one has $a_0 = -\rho_0^2 < 0$. Remarkably, anticorrelations develop even though there is no exclusion. However, the amplitude is different and, unlike in the exclusion process, these anticorrelations vanish in a finite system driven by a boundary gradient as time tends to infinity.

V. PARTICLE NUMBER FLUCTUATIONS

We define as $Q(t) = N(t) - N(0)$ the net number of particles that have entered or left the system until time t. The mean $\langle Q(t) \rangle$ is evaluated using Eqs. (A3), (A4), and (A6) and one gets

$$\langle Q(t) \rangle = \Delta \rho \sum_{k=1}^{\infty} g_k(t) = \Delta \rho e^{-t} \sum_{k=1}^{\infty} (2k-1) I_k(t)$$

$$= \Delta \rho \left[t e^{-t} [I_0(t) + I_1(t)] - \frac{1 - e^{-t} I_0(t)}{2} \right]. \quad (45)$$

The mean grows asymptotically with the power law

$$\langle Q(t) \rangle \sim \Delta \rho \sqrt{\frac{2t}{\pi}} \quad (46)$$

characteristic of diffusive processes.

The variance

$$\sigma^2(t) = \langle Q^2(t) \rangle - \langle Q(t) \rangle^2 \quad (47)$$

may be split into three different parts

$$\sigma^2(t) = \langle N^2(t) \rangle - \langle N(t) \rangle^2 + \langle N^2(0) \rangle - \langle N(0) \rangle^2$$

$$- 2[\langle N(t) N(0) \rangle - \langle N(t) \rangle \langle N(0) \rangle]. \quad (48)$$

Since the initial state is a product state one has $\langle N(t) N(0) \rangle - \langle N(t) \rangle \langle N(0) \rangle = \kappa_0 (d/d\rho_0) \langle N(t) \rangle$, where κ_0 [see Eq. (23)] is the compressibility of the system in the initial state.

On the other hand, because of the exclusion principle, one can write

$$\langle N^2(t) \rangle - \langle N(t) \rangle^2 = -(\Delta \rho)^2 K(t) + \sum_{k=1}^{\infty} [\rho_k(t) - \rho_k^2(t)]$$

$$(49)$$

with the double sum

$$-(\Delta \rho)^2 K(t) \equiv 2 \sum_{k=1}^{\infty} \sum_{l=k+1}^{\infty} C_{k,l}(t). \quad (50)$$

Using

$$\frac{d}{d\rho_0} \langle n_k(t) \rangle = 1 - g_k(t) \quad (51)$$

we rewrite Eq. (48) in the form

$$\sigma^2(t) = \sum_{k=1}^{\infty} [\{\kappa_0 + \kappa^* + (\Delta \rho)^2\} g_k(t) - (\Delta \rho)^2 g_k^2(t)]$$

$$- (\Delta \rho)^2 K(t) \quad (52)$$

convenient for studying its asymptotic behavior. The sum over g_k has been calculated in Eq. (45). The evaluation of the other sums is rather technical, the details are presented in Appendix B. One finds the following asymptotics:
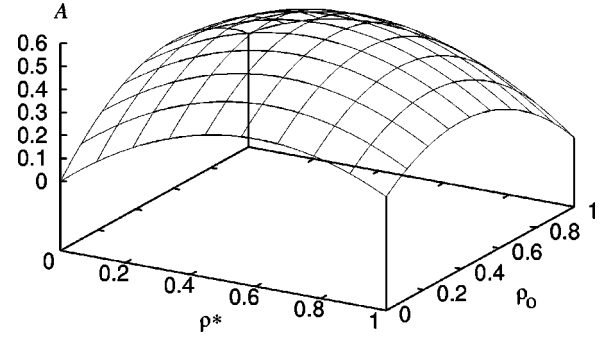

FIG. 2. Amplitude A of the particle number fluctuations as a function of the boundary density ρ^* and the initial bulk density ρ_0.

$$K(t) \sim (3 - 2\sqrt{2}) \sqrt{\frac{4t}{\pi}}, \quad (53)$$

$$\sum_{k=1}^{\infty} g_k^2(t) \sim (\sqrt{2} - 1) \sqrt{\frac{4t}{\pi}}, \quad (54)$$

and therefore

$$\sigma^2(t) = A(\rho_0, \rho^*) \sqrt{t} \quad (55)$$

with

$$A(\rho_0, \rho^*) = \sqrt{\frac{2}{\pi}} [\kappa_0 + \kappa^* + (3 - 2\sqrt{2})(\Delta \rho)^2]. \quad (56)$$

The amplitude A is symmetric under interchange of the macroscopic quantities ρ_0, ρ^* and convex in the physical domain $0 \leq \rho_0, \rho^*, \leq 1$ with a local maximum at $\rho_0 = \rho^* = 1/2$ (Fig. 2). For an initially completely filled lattice ($\rho_0 = 1$) we recover the result presented previously [16]. For the trivial case of noninteracting particles one has $(\sqrt{2} - 1) \rho_0^2$ instead of the gradient term in Eq. (56).

Within the Rubinstein model for reptation [15] and its extension [6] the particle number of the symmetric exclusion process is proportional to the tube length of a polymer and hence proportional to the experimentally accessible visual length of a fluorescence marked entangled macromolecule such as DNA. A nonequilibrium initial density $\rho_0 > \rho^*$ corresponds to a stretched conformation that may be approximated by dragging a molecule through a dense solution with optical tweezers [14]. It has been shown that the relaxing tube length calculated from the symmetric exclusion process is in good agreement with experimental data in the universal initial-time regime [6]. The particle number fluctuations (the lattice sum over the two-point density correlation function) that we obtain yield the evolution of the tube length fluctuations of the polymer chain. For an initially fully stretched polymer ($\rho_0 = 1$) the result, Eqs. (55) and (56), has been discussed in a recent publication [16]. We see here that a partially stretched chain displays qualitatively similar relaxational behavior. In contrast to the Langevin approach used in Rouse-based standard reptation theory [13] where A depends solely on the equilibrium tube length fluctuations, the exclusion model predicts a dependence also on the initial

state (via the compressibility κ_0) and the amount of stretching $\Delta\rho$. These features arise from the static interactions between particles (hard-core, in our case) that are neglected in the purely entropic Rouse model.

VI. CONCLUSIONS

The main results of our study are the exact expression (40) and scaling form (42), respectively, of the time-dependent two-point density correlation function and the asymptotic variance (55) in the number of particles that have left or entered the system up to time t. It turns out that the nonequilibrium behavior of the model is largely determined by the dynamical diffusion length $\tilde{L}$ and by three static macroscopic quantities, viz., the compressibilities κ_0, κ^* of the initial and equilibrium states resp., and the density gradient $\Delta\rho$ between the (equilibrium) boundary and the (initial) bulk density. With regard to polymer reptation this result (56) supports our previous conclusion [16] that standard reptation theory is an oversimplified model of the relaxation process of stretched, entangled polymers.

On a local level we find an inverse relationship between the range $\xi \propto \tilde{L}$ of (anti-)correlations and their strength $\propto 1/\tilde{L}$. It is interesting to quantitatively compare the expression (44) for the correlation function in the boundary region of the system with the *stationary* nonequilibrium correlation function C^* of a finite system of L sites with two different reservoir densities $\rho^- = \rho^*$, $\rho^+ = \rho_0$ as considered in the Sec. II. The external density gradient imposes a stationary current that is given by $\hat{j}^* = \Delta\rho/L$. In terms of the scaling variables $x = k/L$, $y = l/L$ one finds $C^* = -L(\hat{j}^*)^2 x(1-y)$ [Refs. [10,19]] that is of the same form as Eq. (44). This result suggests that the state of the open system in the boundary region (i.e., at distances small compared to the diffusion length $\tilde{L}$) is similar to the nonequilibrium steady state of a finite system of size $L = \tilde{L}$. Hence we can identify three distinct length scales where the system displays different behavior. On the scale of the lattice constant $a = 1$ the system (in the boundary region) is in local equilibrium as is the bulk state of the finite stationary system [10]. On intermediate scales $a \ll r \ll \tilde{L}$ the system is locally (i.e., in the boundary region) stationary, but not in equilibrium. On large scales $r \gg \tilde{L}$ the system is neither in equilibrium nor stationary, but displays relaxational behavior and dynamical scaling.

It is no surprise that the qualitative features of the relaxation process in simple symmetric exclusion can be described in terms of dynamical scaling with the diffusion length $\tilde{L}(t)$ and the universal power law $\sqrt{t}$ characteristic for diffusive dynamics. Yet it is gratifying to have a simple, but nontrivial model, where not only scaling theories can be verified explicitly, but also scaling functions can be calculated. An interesting open problem remains the question whether these results can be obtained from more widely applicable coarse-grained hydrodynamic approaches [10] for stochastic interacting particle systems. A direct experimental study of the questions addressed here appears to be feasible by studying colloidal particles in a setup similar to that used

in [9]. In such an experiment the influence of direct particle interactions in addition to pure hard-core repulsion can be studied.

APPENDIX A: MODIFIED BESSEL FUNCTIONS AND ELLIPTIC INTEGRALS

1. Modified Bessel functions

Here we list some useful properties (see, e.g., [21]) of the modified Bessel functions

$$I_n(t) = \frac{1}{2\pi} \int_{-\pi}^{\pi} d\phi \, e^{i\phi n + \cos\phi t} \qquad (A1)$$

with integer index n.

(i) Asymptotic behavior ($t \to \infty$, $u = n^2/t$ finite)

$$e^{-t} I_n(t) \sim \frac{1}{\sqrt{2\pi t}} e^{-n^2/(2t)}. \qquad (A2)$$

(ii) Recursion relations

$$I_n(t) = I_{-n}(t), \qquad (A3)$$

$$2n I_n(t) = t(I_{n-1}(t) - I_{n+1}(t)), \qquad (A4)$$

$$2\frac{d}{dt} I_n(t) = I_{n-1}(t) + I_{n+1}(t). \qquad (A5)$$

(iii) Summation formulas

$$\sum_{n=-\infty}^{\infty} e^{-t} I_n(t) = 1, \qquad (A6)$$

$$\sum_{n=-\infty}^{\infty} I_n(t) I_{n+m}(t) = I_m(2t). \qquad (A7)$$

(iv) Integrals: For m, n integers, one has

$$I_m(t) I_n(t) = \frac{2}{\pi} \int_0^{\pi/2} d\theta \cos[(n \mp m)\theta] I_{n \pm m}(2t\cos\theta). \qquad (A8)$$

One defines [22] the functions $f_{q,s}(t)$, where q is a positive integer and s is an integer, as

$$f_{q,s}(t) \equiv \sum_{p=0}^{\infty} \binom{p+q-1}{p} 2^q (-1)^p I_{q+s+2p}(t). \qquad (A9)$$

One can also show, using the integral representation of the modified Bessel functions, that for $s \geq 0$, one has

$$f_{q,s}(t) = \frac{1}{(q-1)!} \int_0^t du\, I_s(u)(t-u)^{q-1}. \quad \text{(A10)}$$

The following useful equalities also hold:

$$f_{q,s}(t) = \tfrac{1}{2}[f_{q+1,s+1}(t) + f_{q+1,s-1}(t)], \quad \text{(A11)}$$

$$f_{q+1,1}(t) = f_{q,0}(t) - \frac{t^q}{q!}, \quad \text{(A12)}$$

$$f_{q+1,-1}(t) = f_{q,0}(t) + \frac{t^q}{q!}, \quad \text{(A13)}$$

where the first equality follows from the integral representation of $f_{q,s}(t)$, the second follows from integration by parts of Eq. (A10) and the third follows from the two above.

One can also show [23], that the following identity holds:

$$\sum_{p=0}^{\infty} I_{p+m}(x)y^p = x \int_0^1 dv\, (1-v)^m \exp\left(-\frac{1}{2}xyv^2 + xyv\right)$$

$$\times I_{m-1}(x(1-v)), \quad \text{(A14)}$$

where m is a positive integer.

2. Elliptic integrals

The following relations [24] are used in the calculation of Laplace transforms:

$$\int_0^{\pi/2} d\theta\, \frac{\sin^2\theta}{(1-\alpha^2\sin^2\theta)\sqrt{1-k^2\sin^2\theta}}$$

$$= \frac{\pi(1-\Lambda_0(\phi,k))}{2\sqrt{\alpha^2(1-\alpha^2)(\alpha^2-k^2)}}, \quad \text{(A15)}$$

where $k < \alpha$, $\sin\phi = \sqrt{(1-\alpha^2)/(1-k^2)}$ and $\Lambda_0(\phi,k)$ is given in terms of elliptic functions by

$$\Lambda_0(\phi,k) = \frac{2}{\pi}[E(k)F(\phi,k') + K(k)E(\phi,k')$$

$$- K(k)F(\phi,k')], \quad \text{(A16)}$$

where $K(k)$ and $E(k)$ are the complete elliptic integrals and $F(\phi,k')$, $E(\phi,k')$ are the elliptic integrals of first and second kind, with $k' = \sqrt{1-k^2}$.

APPENDIX B: DERIVATION OF THE EXACT EXPRESSION FOR THE TWO-POINT CORRELATION FUNCTION

In order to derive the exact expression for the two-point correlation function from Eq. (37), one needs, as stated above, to perform the double sum $\Sigma_{k_0=1}^{\infty}\Sigma_{l_0=k_0+1}^{\infty}$ in Eq. (37) and then to determine the contour of integration of the double integral in this equation which will yield the correct

initial condition, namely, $C_{k,l}(0) = 0$. In order to perform the first step, the key point is to realize that one can write the factors $S(p_1,p_2)-1$, $S(-p_1,p_2)-1$ and $S(-p_1,p_2)S(p_1,p_2)-1$ that appear in Eq. (37) in a way such that the sums over l_0 coming from each of these terms can be written as the difference of two sums starting at neighboring arguments, e.g., $l_0 = k_0 + 1$ and $l_0 = k_0 + 2$, and can thus be easily performed using the telescopic property of sums. Furthermore, after some tedious but straightforward algebraic manipulations one can show that the sums over k_0 can also be performed in the same way, i.e., using the telescopic property. When performing this second sum, one also generates one extra term that cancels exactly the first term of Eq. (37). One obtains, after interchanging p_1 and p_2, the following result:

$$C_{k,l}(t) = -2(\Delta\rho)^2 \int' \int' \frac{dp_1}{2\pi} \frac{dp_2}{2\pi} \exp[-(\epsilon_{p_1} + \epsilon_{p_2})t]$$

$$\times \left(\frac{1+e^{-ip_1}}{1-2e^{-ip_1}+e^{-ip_1-ip_2}} e^{-ip_2 k - ip_1 l} \right.$$

$$\left. - \frac{1+e^{-ip_1}}{1-2e^{-ip_1}+e^{-ip_1+ip_2}} e^{-ip_2 k - ip_1 l} \right). \quad \text{(B1)}$$

Now we need to determine the appropriate contour of integration in Eq. (B1). But in fact, one does not need to determine it explicitly. Assuming that the contour includes the origin, one can use the identity $1/x = \int_0^{\infty} d\alpha\, e^{-\alpha x}$ to represent each of the denominators of Eq. (B1) as an integral over α and then formally expand the resulting exponentials under the integration sign in powers of e^{-ip_1} and $e^{-ip_1-ip_2}$ for the first denominator, and e^{-ip_1} and $e^{-ip_1+ip_2}$ for the second denominator. If one then performs the integrals over p_1, p_2, and α, one obtains the following result for $C_{k,l}(t)$:

$$C_{k,l}(t) = -(\Delta\rho)^2 e^{-2t} \sum_{p,q=0}^{\infty} \binom{p+q}{p} 2^{q+1}(-1)^p$$

$$\times [I_{p+k}(t)I_{p+q+l}(t) + I_{p+k}(t)I_{p+q+1+l}(t)$$

$$- I_{p-k}(t)I_{p+q+l}(t) - I_{p-k}(t)I_{p+q+1+l}(t)].$$

$$\text{(B2)}$$

Notice that the existence of the expansions depends on the convergence of the resulting series, which implicitly fixes the contour. It can be easily checked from the properties of the modified Bessel functions that this expression does indeed obey the initial condition $C_{k,l}(0) = 0$ (notice that $k < l$).

This expression is still rather cumbersome to use. If we apply (A8) to products of two modified Bessel functions, we will obtain, with $m = l - k$, $n = l + k$, $r = q + 2p$

$$C_{k,l}(t) = -\frac{2(\Delta\rho)^2}{\pi} e^{-2t} \int_0^{\pi/2} d\theta \sum_{p,q=0}^{\infty} \binom{p+q}{p} 2^{q+1}(-1)^p$$

$$\times \{I_{r+1+(n-1)}(2t\cos\theta)\cos[(q+m)\theta]$$

$$+ I_{r+1+n}(2t\cos\theta)\cos[(q+1+m)\theta]$$

$$- I_{r+1+(m-1)}(2t\cos\theta)\cos[(q+n)\theta]$$

$$- I_{r+1+m}(2t\cos\theta)\cos[(q+1+n)\theta]\}. \tag{B3}$$

One can now write (B3) in terms of the $f_{q,s}(t)$ functions that were defined above. One has

$$C_{k,l}(t) = -\frac{2(\Delta\rho)^2}{\pi} e^{-2t}$$

$$\times \int_0^{\pi/2} d\theta \sum_{q=0}^{\infty} \{f_{q+1,l+k-1}(2t\cos\theta)$$

$$\times \cos[(q+l-k)\theta] + f_{q+1,l+k}(2t\cos\theta)$$

$$\times \cos[(q+l-k+1)\theta]$$

$$- f_{q+1,l-k-1}(2t\cos\theta)\cos[(q+l+k)\theta]$$

$$- f_{q+1,l-k}(2t\cos\theta)\cos[(q+l+k+1)\theta]\}. \tag{B4}$$

Using the integral representation (A10) and summing over q one obtains, after the substitution $u = 2(t-v)\cos\theta$ in the integral over u, the solution (38) where the functions $F_{m,n}(x)$ are given by Eq. (39).

Since the steps that led from Eq. (B1) to Eq. (B2) are only formal, one should check explicitly that Eq. (38) is indeed a solution of the equations of motion for the joint probabilities $\langle n_k(t)n_l(t)\rangle$. This is trivial for $l \neq k+1$. For $l = k+1$ one can show that the unphysical amplitudes $C_{k,k}(t)$, $C_{k+1,k+1}(t)$ that are generated by the time derivative of $\langle n_k(t)n_{k+1}(t)\rangle$, obey the following identity:

$$C_{k,k}(t) + C_{k+1,k+1}(t) - 2C_{k,k+1}(t)$$

$$= -(\Delta\rho)^2 e^{-2t}[I_k(t) + I_{k+1}(t)]^2, \tag{B5}$$

which cancels exactly the unphysical contribution coming from the term $[\rho_k(t) - \rho_{k+1}(t)]^2$ [see Eqs. (27) and (28)] that also appears in the equation for $\langle n_k(t)n_{k+1}(t)\rangle$, thus showing that $\langle n_k(t)n_{k+1}(t)\rangle$ obeys the correct equation of motion. This identity can be proved by considering the expression for the left-hand side of Eq. (B5) as given in terms of the integral representation (38). After some cancellations between the terms, one uses the identity

$$\frac{d}{dv}(\exp\{v[1+\cos(2\theta)]\}\cos[v\sin(2\theta)+s\theta])$$

$$= 2\exp\{v[1+\cos(2\theta)]\}\cos[v\sin(2\theta)+(s+1)\theta] \tag{B6}$$

to integrate the resulting expression by parts. Applying the identity (A8) to the remaining integrals, in order to transform

them into products of modified Bessel functions yields the desired result. An alternative route to derive Eq. (B5) is to use the representation (B4) and the equalities (A11) to (A13).

APPENDIX C: ASYMPTOTICS OF σ^2

1. Sum over $g_k^2(t)$

This expression arises in the summation over $\langle n_k(t)\rangle^2$ that forms part of the "dynamical compressibility" $\langle N^2(t)\rangle - \langle N(t)\rangle^2$ entering the expression for $\sigma^2(t)$. It is convenient to split this sum into three different parts. One has

$$\sum_{k=1}^{\infty} g_k^2(t) = -e^{-2t}\sum_{k=1}^{\infty} I_k^2(t) - 2e^{-2t}\left(\sum_{k=1}^{\infty} I_k(t)\right)^2$$

$$+ 4e^{-2t}\sum_{k=1}^{\infty}\sum_{p=k}^{\infty}\sum_{q=k}^{\infty} I_p(t)I_q(t). \tag{C1}$$

The first part can be evaluated by using Eqs. (A3) and (A7), the second part by using Eq. (A6). To evaluate the third part we rewrite the summation as

$$\sum_{k=1}^{\infty}\sum_{p=k}^{\infty}\sum_{q=k}^{\infty} I_p(t)I_q(t) = \sum_{k=1}^{\infty}\left(k\,I_k^2(t) + 2\sum_{p=0}^{k-1} p\,I_p(t)I_k(t)\right) \tag{C2}$$

and apply Eqs. (A3), (A4), (A6), and (A7). One obtains

$$\sum_{k=1}^{\infty}\sum_{p=k}^{\infty}\sum_{q=k}^{\infty} I_p(t)I_q(t) = \frac{t}{2}[e^t\{I_0(t) + I_1(t)\}$$

$$- \{I_0(2t) + I_1(2t)\}]. \tag{C3}$$

Putting everything together yields

$$\sum_{k=1}^{\infty} g_k^2(t) = 2t[e^{-t}[I_0(t) + I_1(t)] - e^{-2t}[I_0(2t) + I_1(2t)]]$$

$$- [1 - 2e^{-t}I_0(t) + e^{-2t}I_0(2t)]/2. \tag{C4}$$

The second part of this expression is subleading in time and can be ignored in the study of the asymptotic behavior.

2. Laplace transform of $K(t)$

In order to perform the sum on the right-hand side of Eq. (50), we consider the Laplace transform of the correlation function $C_{k,l}(t)$ as given by Eq. (38). Since the expression (38) involves a convolution of two functions $\int_0^t dv\,f(t-v)g(v)$, such transformation simplifies considerably the calculations, because the Laplace transform of such a convolution is the product of the Laplace transforms of the two functions. Writing the Laplace transform as $\tilde{C}_{k,l}(s)$, one has

$$\widetilde{C}_{k,l}(s) = -\frac{4(\Delta\rho)^2}{\pi}\int_0^{\pi/2}d\theta\frac{\cos\theta}{\sqrt{(s+2)^2-4\cos^2\theta}}$$

$$\times\frac{1}{(s+1)^2-2(s+1)\cos(2\theta)+1}[\mathcal{F}_{k+l-1,l-k}(s)$$

$$+\mathcal{F}_{k+l,l-k+1}(s)-\mathcal{F}_{l-k-1,k+l}(s)-\mathcal{F}_{l-k,k+l+1}(s)],$$
$$\text{(C5)}$$

where the functions $\mathcal{F}_{m,n}(s)$ are defined by

$$\mathcal{F}_{m,n}(s)=\left(\frac{s+2-\sqrt{(s+2)^2-4\cos^2\theta}}{2\cos\theta}\right)^m[(s+1)\cos(n\theta)$$

$$-\cos\{(n-2)\theta\}]. \qquad\text{(C6)}$$

An additional advantage of (C5) with respect to Eq. (38) is that the sums over k and l now reduce to summing two geometric series, due to the form of $\mathcal{F}_{m,n}(s)$. Performing such sums, one obtains after computing some standard integrals using the residue theorem, the following expression for the Laplace transform $\widetilde{K}(s)$ of $K(t)$:

$$\widetilde{K}(s)=-\frac{2}{s^{3/2}}\left(\sqrt{s+2}-\frac{\sqrt{s+4}}{4}\right)+\frac{3}{2s}+\frac{4(3s+4)}{\pi(s+2)^3}$$

$$\times\int_0^{\pi/2}d\theta\frac{\sin^2\theta}{[1-\alpha^2(s)\sin^2\theta]\sqrt{1-k^2(s)\sin^2\theta}}$$

$$-\frac{8}{\pi(s+2)^2}$$

$$\times\int_0^{\pi/2}d\theta\frac{\sin^2\theta}{[1-\alpha'^2(s)\sin^2\theta]\sqrt{1-k^2(s)\sin^2\theta}},$$
$$\text{(C7)}$$

where $\alpha^2(s)=4(s+1)/(s+2)^2$, $\alpha'^2(s)=2/(s+2)$, and $k^2(s)=4/(s+2)^2$. Since the last two terms are of the form (A15), one can now expand the elliptic integrals at small s [24]. The most singular terms of this expansion, i.e., the terms that diverge as $1/s^{3/2}$ at small s diverge like $\sqrt{t}$ at large t in the time-domain, i.e., after inverting the Laplace transformation. Collecting all these leading-order terms in the expansion of $\widetilde{K}(s)$ yields the result given in Eq. (53).

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