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Calculating exact propagators in single-file systems via the reflection principle

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The dynamics of tagged particles in diffusive single-file systems (one-dimensional systems where the particles are not able to pass each other) is investigated. The presented approach, based on the reflection principle, yields exact propagators for quite a general class of systems. Examples are considered explicitly and compared both with results of computer simulations and, in one case, with the asymptotic behavior known from the literature. Practical implications of the results to the interpretation of data from scattering experiments and to the application of single files for the controlled release of particles are discussed. [S1063-651X(98)02604-X]

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I. INTRODUCTION

One-dimensional diffusion of a set of particles that are not able to change their order is known as *single-file diffusion*. Quite a number of systems in different fields of science show such a behavior, e.g., superionic or organic conducters [1], ion channels through biological membranes [2], or many zeolites with a one-dimensional channel system (Mordenite, L, $AlPO_4$-5, etc.) [3]. While the collective motion of the particles in such a single-file system proceeds just like that of independent particles, the dynamics of *tagged particles* is considerably different [4,5]. Its analytical treatment proves complicated because the motions of all individual particles are correlated to each other over the entire system [6]. Most theoretical investigations in the literature are, for this reason, restricted to mean quantities or limiting cases. In [7], a formalism is developed yielding the *propagator* of a tagged pointlike particle in a single-file system on the basis of the propagator of an isolated (i.e., noninteracting) particle in the same system. Due to an approximation partly neglecting the aforementioned correlations, the result is only valid for sufficiently large times. A number of interesting phenomena, e.g., the transition from the initial Fickian behavior to single-file diffusion, or the diffusion of tagged particles in finite single-file systems, however, involve rather short times not covered by the range of validity of [7].

The formalism presented in this paper is free of approximations, i.e., it considers the correlations completely, and therefore yields exact propagators for all observation times. It may be applied to a wide class of single-file systems, including systems without translational invariance or with inhomogeneous initial particle densities. Generally, the particles are assumed to be pointlike and diffusing in a continuous one-dimensional space.

In order to illustrate the application of the general formulas, special systems will be considered at certain stages of the calculation. Two examples are, in addition, of practical relevance.

Example 1. Many experimentally investigated single-file systems may be idealized as *infinite, homogeneously occupied channels*. As powerful tools, quasielastic neutron scattering [8] and pulsed field gradient NMR [9,10] are widely used in this field. The interpretation of their data relies on the assumption that the propagator of tagged particles is a Gaussian [11,3]. For large observation times, the validity of this assumption has been shown analytically [7,12]. Some experiments, such as neutron scattering at zeolitic single-file systems [13], however, are able to resolve rather short observation times, belonging to the aforementioned transition region between normal and single-file diffusion where exact propagators have not yet been obtained. This paper will close this gap.

Example 2. Applications such as controlled-release systems [14] require systems that release particles in a slow, well-defined way. In single-file systems, tracer processes are much slower than in systems undergoing normal diffusion due to the strong mutual hindrance of the particles [15]. Moreover, the particles leave the channel in exactly the same order as they entered it previously. As a basis of a quantitative analysis of the release, we will consider the following process. Initially, the finite single-file channel is in sorption equilibrium with an infinite external reservoir of particles so that the channel is homogeneously occupied with a given particle concentration c. At time zero, the channel openings are exposed to vacuum, whence the particles will, one after another, be desorbed from the file without any chance to return. By means of the calculation presented in this paper, an individual of these particles starting at an arbitrary given position within such a *finite channel with absorbing boundaries* can be traced.

The motion of particles in a single-file system is determined by two kinds of influences: first, the particle-channel interaction, which acts on each individual particle causing it to move, and second, the particle-particle interaction, which inhibits the particles from changing their order. Section II, as a preliminary, is devoted to the first kind of influence, thus considering an *isolated particle* in the system.

Prepared in this way, Sec. III turns to a single-file system occupied by a *finite number of particles* that influence each other via the particle-particle interaction. Three steps of calculation lead to a general expression of the propagator of a tagged particle, which is subsequently applied to special single-file systems.

The aforementioned examples, however, involve an *infi-*

nite number of particles. Thus, as a fourth calculation step, Sec. IV considers this limit. After deriving the general propagator, the examples are explicitly treated and discussed. The Conclusion indicates possible generalizations.

II. PRELIMINARY: THE ISOLATED PARTICLE

A. The particle-channel interaction

The interaction between each individual particle and the channel is described by the *isolated-particle propagator* $f(x|a)$. Generally, the presented formalism assumes that this propagator represents the solution of a partial differential equation that follows from the underlying model of diffusion. The variable x denotes the position of the isolated particle at time t, while a is the initial position at time 0. The propagator represents the conditional probability density of the particle position, i.e., $f(x|a)dx$ gives the propability that the particle is to be found between x and $x+dx$ under the condition that it started at a. [If needed, the time argument will be written as a superscript $f^t(x|a)$, but usually it will, for lucidity, be omitted.] The propagator is a normalized distribution, i.e.,

$$\int_{-\infty}^{\infty} f(x|a)dx = 1. \tag{1}$$

In addition to the probability density $f(x|a)$, the corresponding distribution function

$$g(x|a) := \int_{-\infty}^{x} f(\xi|a)d\xi \tag{2}$$

will be required. In the following, several well-known relations concerning the dynamics of an isolated particle in some special systems are listed.

B. Example: The infinite channel

The most common differential equation describing diffusional processes is Fick's second law

$$\dot{f}(x|a) = D\frac{\partial^2 f}{\partial x^2}(x|a), \tag{3}$$

known as the diffusion equation. If the particle starts at position a one has the initial condition

$$f^0(x|a) = \delta(x-a). \tag{4}$$

The solution for an infinite channel is given by

$$f(x|a) = f_i(x|a) = \Phi(x-a), \tag{5}$$

where $\Phi(u)$ is a shorthand for the Gaussian distribution given explicitly in Eq. (A1) in the Appendix. The corresponding distribution function reads, according to Eqs. (2) and (A4),

$$g_i(x|a) = \Gamma(x-a), \tag{6}$$

where the function $\Gamma(x)$ is defined in Eq. (A2).

We remark that Eq. (3) is by no means a unique choice of a differential equation modeling diffusion. Instead, one might make use of the telegrapher's equation

$$\frac{D}{\sigma^2}\ddot{f}(x|a) + \dot{f}(x|a) = D\frac{\partial^2 f}{\partial x^2}(x|a), \tag{7}$$

which takes into account that, for very short times t, the particle actually moves ballistically rather than diffusively. The parameter σ can be interpreted as the finite velocity of the particle [16]. For larger times, this equation coincides with the diffusion equation.

C. Example: The finite channel with reflecting boundaries

Now consider a finite channel of length L, extending from $x_L=0$ to $x_R=L$. At both boundaries, the file can be plugged up: The particle will be reflected back on reaching them. Consequently, the probability fluxes through the boundaries disappear. Since, according to Fick's first law, the probability flux is proportional to the first derivative of the propagator, this yields the well-known boundary conditions

$$\frac{\partial f_{rr}}{\partial x}(0|a) = 0, \quad \frac{\partial f_{rr}}{\partial x}(L|a) = 0. \tag{8}$$

The propagator for this system is obtained as a solution of the diffusion equation (3) subject to these boundary conditions. This solution can easily be obtained from that of the infinite system (5) via the well-known reflection principle [17,10]. If only one reflecting boundary were present, say at the left boundary, the solution would have to be reflected and added, while the density beyond the boundary would have to be set zero:

$$f_r(x|a) = \begin{cases} \Phi(x-a)+\Phi(x+a), & x \geqslant 0 \\ 0, & x<0. \end{cases} \tag{9}$$

In the presence of two reflecting boundaries, each boundary, in effect, ''reflects'' the other boundary as well, resulting in an infinite number of ''true'' or ''reflected'' boundaries and therefore in an infinite number of reflected solutions ($0 \leqslant x \leqslant L$, $0 \leqslant a \leqslant L$):

$$f_r(x|a) = \sum_{k=-\infty}^{\infty} [\Phi(x-a+2kL)+\Phi(x+a+2kL)]. \tag{10}$$

The corresponding distribution function follows as

$$g_r(x|a) = \sum_{k=-\infty}^{\infty} [\Gamma(x-a+2kL)-\Gamma(-a+2kL) + \Gamma(x+a+2kL)-\Gamma(a+2kL)]. \tag{11}$$

D. Example: The finite channel with absorbing boundaries

In contrast to Sec. II C, any particle reaching one of the boundaries is now assumed to be absorbed immediately into the surrounding space from which it will never return into the channel. This implies that the probability of finding the isolated particle at one of the boundaries vanishes, whence the boundary conditions read

$$f_{aa}(0|a)=0, \quad f_{aa}(L|a)=0. \tag{12}$$

The propagator will only be required for $0\leqslant x\leqslant L$, i.e., for positions inside the channel. In this region, it is given by

$$f_{aa}(x|a)=\begin{cases}\sum_{k=-\infty}^{\infty}[\Phi(x-a+2kL)-\Phi(x+a+2kL)], & 0\leqslant a\leqslant L\\ 0, & L<a.\end{cases} \tag{13}$$

(In the case $0\leqslant a\leqslant L$ where the isolated particle starts at an arbitrary position within the channel, the propagator is, as in Sec. II C, obtained via the reflection principle, though by antireflection, i.e., subtracting rather than adding the reflected solutions. In the case $a>L$ where the particle initially is already situated to the right of the right boundary, i.e., outside the channel, the probability of finding the particle at any position within the channel vanishes because we assumed that it cannot enter the channel from outside. The missing case $a<0$ will not be required in the latter calculation.)

In order to obtain the corresponding distribution function one needs the probability that the particle is situated to the left of the left boundary, i.e., it has been desorbed at this side of the channel,

$$\int_{-\infty}^{0} f_{aa}(x|a)dx=\begin{cases}2\sum_{k=0}^{\infty}[\Gamma(2L-a+2kL)-\Gamma(a+2kL)], & 0\leqslant a\leqslant L\\ 0, & L<a.\end{cases} \tag{14}$$

(This expression follows from the time integral over the flux out of the left boundary.) Then the distribution function can be calculated via its definition by Eq. (2):

$$g_{aa}(x|a)=\begin{cases}\sum_{k=0}^{\infty}[\Gamma(x+2L-a+2kL)+\Gamma(-x+2L-a+2kL)\\ \quad -\Gamma(x+a+2kL)-\Gamma(-x+a+2kL)], & 0\leqslant a\leqslant L\\ 0, & L<a.\end{cases} \tag{15}$$

III. THE SINGLE-FILE SYSTEM WITH FINITELY MANY PARTICLES

A. First step: The total propagator

In Sec. II a system with an isolated pointlike particle in a diffusional channel was considered. The state of this system was given by the coordinate x of the particle and the dynamics could be described by the propagator $f(x|a)$ characterizing the particle-channel interaction including the driving diffusion mechanism.

If *several* pointlike particles diffuse in the same channel the state may be described by the vector $\mathbf{x}=(\ldots,x_i,\ldots)$ of the coordinates x_i of the individual particles. In the case of *free* particles (i.e., without particle-particle interaction) the coordinates x_i are statistically independent and the total propagator (i.e., the propagator of the system as a whole) is given by

$$F(\mathbf{x}|\mathbf{a})=\prod_i f(x_i|a_i). \tag{16}$$

This expression can be interpreted as the solution of the differential equation of the isolated particle in the higher-dimensional state space S of the vectors $\mathbf{x}$, where boundaries of the channel, if any, cause corresponding boundary planes perpendicular to the axes of S.

The single-file system is characterized by the impossibility that two particles change places so that the order of the coordinates x_i is strictly preserved during all the dynamical development of the system. This implies that only a certain section S' of the space S is accessible: If the initial order of two arbitrary coordinates is $a_i<a_j$, all points with $x_i>x_j$ are forbidden. Thus the accessible section S' of the state space is demarcated by the planes $x_i=x_j$. The single-file property can therefore be expressed as the condition that the probability flux through these planes vanishes, i.e., these planes act as reflecting boundaries. This means that the total propagator $P(\mathbf{x}|\mathbf{a})$ of the single-file system can be obtained as the solution of the higher-dimensional differential equation subject to the (additional) boundary conditions

$$\left.\frac{\partial P(\mathbf{x}|\mathbf{a})}{\partial n}\right|_{x_i=x_j}=0 \quad \forall i\neq j \tag{17}$$

(derivative normal to the plane $x_i=x_j$), which can be transformed to

$$\left.\left(\frac{\partial P(\mathbf{x}|\mathbf{a})}{\partial x_i}-\frac{\partial P(\mathbf{x}|\mathbf{a})}{\partial x_j}\right)\right|_{x_i=x_j}=0 \quad \forall i\neq j. \tag{18}$$

As was already done in the case of the channel with reflecting boundaries, this boundary condition can be satisfied using the *reflection principle*: If the solution is reflected at a plane and added to the original solution, the derivative of the sum normal to this plane vanishes. If several reflection planes are present, all the reflected planes act as reflection planes as well and one has to consider all possible combinations of reflections.

For the present problem, reflection at a plane $x_i = x_j$ simply means exchanging the coordinates x_i and x_j. The possible combinations of reflections are given by the permutations of the coordinates, so that

$$P(\mathbf{x}|\mathbf{a}) = \begin{cases} \sum_{\pi} F(\mathbf{V}_\pi \mathbf{x}|\mathbf{a}), & x \in S' \\ 0, & x \notin S', \end{cases} \tag{19}$$

where the sum is extended over all permutations π and $\mathbf{V}_\pi$ is the corresponding matrix exchanging the coordinates.

Proof. Consider an arbitrary pair of permutations

$$\pi_1 = \begin{pmatrix} & k & & l & \\ \dots, & x_i, & \dots, & x_j, & \dots \end{pmatrix}, \quad \pi_2 = \begin{pmatrix} & k & & l & \\ \dots, & x_j, & \dots, & x_i, & \dots \end{pmatrix},$$

i.e., in π_1 the coordinate x_i holds the place of coordinate x_k, the coordinate x_j holds the place of coordinate x_l, and π_2 is equal except for the exchange of coordinates x_i and x_j. Now differentiate $P(\mathbf{x}|\mathbf{a})$ with respect to x_i or x_j, respectively, and take the result at an arbitrary point $\mathbf{y}$ (only the terms belonging to the permutations π_1 and π_2, respectively, are written):

$$\left.\frac{\partial P(\mathbf{x}|\mathbf{a})}{\partial x_i}\right|_{\mathbf{x}=\mathbf{y}} = \dots + \left.\frac{\partial F(\mathbf{x}|\mathbf{a})}{\partial x_k}\right|_{\dots, x_k = y_i, \dots, x_l = y_j, \dots} + \dots + \left.\frac{\partial F(\mathbf{x}|\mathbf{a})}{\partial x_l}\right|_{\dots, x_k = y_j, \dots, x_l = y_i, \dots} + \dots,$$

$$\left.\frac{\partial P(\mathbf{x}|\mathbf{a})}{\partial x_i}\right|_{\mathbf{x}=\mathbf{y}} = \dots + \left.\frac{\partial F(\mathbf{x}|\mathbf{a})}{\partial x_l}\right|_{\dots, x_k = y_i, \dots, x_l = y_j, \dots} + \dots + \left.\frac{\partial F(\mathbf{x}|\mathbf{a})}{\partial x_k}\right|_{\dots, x_k = y_j, \dots, x_l = y_i, \dots} + \dots.$$

If $y_i = y_j$ both expressions are equal, whence $P(\mathbf{x}|\mathbf{a})$ satisfies the boundary condition (18) for *any* function $F(\mathbf{x}|\mathbf{a})$.

$P(\mathbf{x}|\mathbf{a})$ can be expressed in a way more convenient to the further calculations by permutating the initial coordinates $\mathbf{a}$ rather than $\mathbf{x}$:

$$P(\mathbf{x}|\mathbf{a}) = \begin{cases} \sum_{\pi} F(\mathbf{x}|\mathbf{V}_\pi \mathbf{a}), & x \in S' \\ 0, & x \notin S'. \end{cases} \tag{20}$$

Proof. The dependence of $F(\mathbf{x}|\mathbf{a})$ on the individual coordinates x_i differs only in the parameters a_i. Since exchanging two coordinates means exchanging the individuality of the dependence on these coordinates it is completely equivalent to exchanging the values of the corresponding components of $\mathbf{a}$.

We remark that the planes $x_i = x_j$ divide the state space S into a large number of disjunct sections. All these sections are geometrically congruent: They can be mapped onto each other by reflection at their demarcating planes. Each of these sections correponds to exactly one order of the coordinates of the particles. This implies that the initial order determines which of these sections represents the accessible section S' [18].

B. Second step: The tagged particle

Let us now consider an individual tagged particle, chosen out of the identical particles of the single-file system. It is labeled by the index 0, i.e., its coordinate is x_0. There are R_L left neighbors with the coordinates $x_{-R_L}, \dots, x_{-1}$ and R_R right neighbors with the coordinates $x_1, \dots, x_{R_R}$. The order of the coordinates is

$$-\infty \leqslant x_{-R_L} < x_{-R_L+1} < \dots < x_{-1} < x_0 < x_1 < \dots < x_{R_R-1} < x_{R_R} \leqslant \infty. \tag{21}$$

The propagator of the tagged particle is obtained from the total propagator by integration over all other coordinates,

$$p(x_0|\mathbf{a}) = \int_{-\infty}^{\infty} dx_{-1} \int_{-\infty}^{\infty} dx_{-2} \cdots \int_{-\infty}^{\infty} dx_1 \int_{-\infty}^{\infty} dx_2 \cdots P(\mathbf{x}|\mathbf{a}). \tag{22}$$

Now Eq. (20) is inserted, whence the range of integration is limited over the accessible space section S corresponding to the order (21):

$$p(x_0|\mathbf{a}) = \int_{-\infty}^{x_0} dx_{-1} \int_{-\infty}^{x_{-1}} dx_{-2} \cdots \times \int_{x_0}^{\infty} dx_1 \int_{x_1}^{\infty} dx_2 \cdots \sum_{\pi} F(\mathbf{x}|\mathbf{V}_\pi \mathbf{a}). \tag{23}$$

This expression, however, cannot be further evaluated. To get a simpler expression one can invoke the fact that, from the point of view of the tagged particle, all its left neighbors are indistinguishable from each other, as are the right neighbors. The accessible region of the tagged particle is limited by the positions of its two next neighbors, irrespective of which individual particles occupy these positions. This implies that the propagator of the tagged particle does not change if both the left and right neighbors are allowed to mutually change their order. Only exchanges with the tagged particle have to remain excluded. Thus one can do with the much weaker condition

$$-\infty \leqslant x_{-R_L} < x_0, \dots, -\infty \leqslant x_{-1} < x_0, \quad x_0 < x_1 \leqslant \infty, \dots, x_0 < x_{R_R} \leqslant \infty \tag{24}$$

and extend the range of integration over the modified accessible space section S'' corresponding to this condition. Mathematically, this is possible because of the congruence of the space sections belonging to different orders of the coordinates. For compensation, however, a normalization Z has to be introduced. If, in addition, Eq. (16) is inserted, one arrives at

$$p(x_0|\mathbf{a})=\frac{1}{Z}\sum_{\pi}\int_{-\infty}^{x_0}dx_{-R_L}\cdots\int_{-\infty}^{x_0}dx_{-1}\times\int_{x_0}^{\infty}dx_1\cdots\int_{x_0}^{\infty}dx_{R_R}\prod_i f(x_i|(\mathbf{V}_\pi\mathbf{a})_i). \quad (25)$$

Consider an arbitrary term of this sum (corresponding to a given permutation π) and abbreviate the permutated initial vector by $\mathbf{b}:=\mathbf{V}_\pi\mathbf{a}$. One observes that each integral applies to one of the functions $f(x_i|(\mathbf{V}_\pi\mathbf{a})_i)$ only and that one of the functions remains without integration. According to the sign of the index i, this results, therefore, in three types of factors making up the considered term,

$$\int_{-\infty}^{x_0}f(x_i|b_i)dx_i \quad (i<0, L\text{-type factor}), \quad (26)$$

$$\int_{x_0}^{\infty}f(x_i|b_i)dx_i \quad (i>0,\ R\text{-type factor}), \quad (27)$$

$$f(x_0|b_0) \quad (i=0,\ 0\text{-type factor}). \quad (28)$$

[In the following, the notation ''factor'' will consistently refer to these very factors of the terms of Eq. (25).] Using the distribution function defined in Eq. (2), one can write the L-type factor in the form $g(x_0|b_i)$ and the R-type factor due to Eqs. (1) and (26) in the form $[1-g(x_0|b_i)]$. The term of Eq. (25) considered can thus be written as

$$T(x_0|\mathbf{b})=g(x_0|b_{-R_L})\cdots g(x_0|b_{-1})f(x_0|b_0)\times[1-g(x_0|b_1)]\cdots[1-g(x_0|b_{R_R})]. \quad (29)$$

The normalization Z is obtained from the condition

$$\int_{-\infty}^{\infty}p(x_0|\mathbf{a})dx_0=1, \quad (30)$$

which yields via Eqs. (25) and (29)

$$Z=\int_{-\infty}^{\infty}\sum_{\pi}T(x_0|\mathbf{V}_\pi\mathbf{a})dx_0. \quad (31)$$

We define functions

$$g_i(x_0):=g(x_0|a_i), \quad (32)$$

which give

$$g_i{}'(x_0)=f(x_0|a_i). \quad (33)$$

If Eq. (29) is multiplied out the terms $T(x_0|\mathbf{V}_\pi\mathbf{a})$ can be written as sums of subterms, each of these subterms consisting of h factors ($R_L+1\leqslant h\leqslant R_L+1+R_R$), where $h-1$ factors are of the form $g_i(x_0)$ and one factor is of the form $g_i{}'(x_0)$. The sign of the subterm is given by

$$(-1)^{h-R_L-1}. \quad (34)$$

Now we group all the subterms of Eq. (31) (i.e., *all* subterms of *all* terms T) in such a way that every group contains exactly h subterms, each of which consisting of exactly h factors; in a given group, the occurring indices i_k ($k=1,\dots,h$) of the factors of the subterms belong to the same subset of the set of possible indices; and the subterms of a given group differ in the index of the factor $g_i{}'(x_0)$. Then the integral over such a group can be found via the identity

$$\int_{-\infty}^{\infty}(g_{i_1}'g_{i_2}\cdots g_{i_h}+g_{i_1}g_{i_2}'\cdots g_{i_h}+\cdots+g_{i_1}g_{i_2}\cdots g_{i_h}')dx_0=[g_{i_1}g_{i_2}\cdots g_{i_h}]_{-\infty}^{\infty}=1, \quad (35)$$

which follows from $g_i(-\infty)=0$ and $g_i(\infty)=1$ due to Eqs. (1) and (2). This means that the value of the integral over a group is equal to the (common) sign (34) of its subterms and therefore depends only on h. For each h, there are $\binom{R_R}{h-R_L-1}$ $(R_L+1+R_R)!$ subterms ([R_L+1+R_R]! is the number of all terms and $\binom{R_R}{h-R_L-1}$ is the number of subterms per term with h factors because it is the number of possibilities to choose the $(h-R_L-1)$ R-type factors contributing with $g_i(x_0)$ rather than with 1). These subterms are parted into groups of h members each, whence one gets

$$Z=\sum_{h=R_L+1}^{R_L+1+R_R}(-1)^{h-R_L-1}\frac{\binom{R_R}{h-R_L-1}(R_L+1+R_R)!}{h}=(R_L+1+R_R)!\sum_{k=0}^{R_R}\frac{(-1)^k}{R_L+1+k}\binom{R_R}{k}. \quad (36)$$

The sum can be done analytically [19] and gives

$$Z=R_L!R_R!. \quad (37)$$

This result allows an illustrative interpretation: Since we have used the modified order (24) of the allowed coordinates rather than the true order (21), we have increased the size of the accessible section of the state space. While the original section S' corresponds to only one permutation of the coordinates, the modified section S'' corresponds to a large number of permutations. Thus the increased section is as many times larger than the original one as the number of permutations allowed by the modified order. The obtained normalization Z gives exactly this number.

C. Third step: The propagator of the tagged particle

The propagator $p(x_0|\mathbf{a})$ is valid for the initital condition $\mathbf{a}$, fixing explicitly the initial positions of all particles. Now we assume that the left neighbors of the tagged particle initially were randomly distributed over the interval $-\infty\leqslant a_j\leqslant a_0$. The probability density of the initial coordinate a_j of any individual particle is $\varrho_L(a_j|a_0)$, which shall be common to all left neighbors. Likewise, the right neighbors shall be distributed over $a_0\leqslant a_j\leqslant\infty$ according to $\varrho_R(a_j|a_0)$. Of course, the densities ϱ_L and ϱ_R have to be normalized:

$$\int_{-\infty}^{a_0} \varrho_L(a|a_0)da = \int_{a_0}^{\infty} \varrho_R(a|a_0)da = 1. \quad (38)$$

Taking the mean of the propagator over all possible initital conditions according to these densities, one obtains

$$p(x_0|a_0) = \int_{-\infty}^{a_0} da_{-R_L} \cdots \int_{-\infty}^{a_0} da_{-1} \int_{a_0}^{\infty} da_1 \cdots \times \int_{a_0}^{\infty} da_{R_R} \varrho_L(a_{-R_L}|a_0) \cdots \times \varrho_L(a_{-1}|a_0)\varrho_R(a_1|a_0) \cdots \varrho_R(a_{R_R}|a_0) p(x_0|\mathbf{a}). \quad (39)$$

When this mean is performed with the individual terms $T(x_0|\mathbf{V}_\pi\mathbf{a})$ of $p(x_0|\mathbf{a})$, there is a situation analogous to that observed on discussing Eq. (25): Again each integral of Eq. (39) applies to only one factor of the terms $T(x_0|\mathbf{V}_\pi\mathbf{a})$; there are two different types of integrals according to the sign of the index j of the initial coordinate,

$$\int_{-\infty}^{a_0} \cdots \varrho_L(a_j|a_0)da_j \quad (j<0,\ L\text{-type operation}), \quad (40)$$

$$\int_{a_0}^{\infty} \cdots \varrho_R(a_j|a_0)da_j \quad (j>0,\ R\text{-type operation}), \quad (41)$$

and one factor of the term remains without integration (0-type operation). The respective permutation π determines which of the operations is carried out on which of the factors of the term $T(x_0|\mathbf{V}_\pi\mathbf{a})$. Since every combination is possible, nine types of quantities occur as the factors of the resulting terms $\overline{T}_\pi(x_0|a_0)$ of $p(x_0|a_0)$ (where the first index gives the type of factor and the second index the type of operation):

$$k_{LL} = k_{LL}(x_0|a_0) = \int_{-\infty}^{a_0} g(x_0|a)\varrho_L(a|a_0)da,$$

$$k_{0L} = k_{0L}(x_0|a_0) = \int_{-\infty}^{a_0} f(x_0|a)\varrho_L(a|a_0)da,$$

$$k_{RL} = k_{RL}(x_0|a_0) = \int_{-\infty}^{a_0} [1-g(x_0|a)]\varrho_L(a|a_0)da,$$

$$k_{L0} = k_{L0}(x_0|a_0) = g(x_0|a_0),$$

$$k_{00} = k_{00}(x_0|a_0) = f(x_0|a_0), \quad (42)$$

$$k_{R0} = k_{R0}(x_0|a_0) = [1-g(x_0|a_0)],$$

$$k_{LR} = k_{LR}(x_0|a_0) = \int_{a_0}^{\infty} g(x_0|a)\varrho_R(a|a_0)da,$$

$$k_{0R} = k_{0R}(x_0|a_0) = \int_{a_0}^{\infty} f(x_0|a)\varrho_R(a|a_0)da,$$

$$k_{RR} = k_{RR}(x_0|a_0) = \int_{a_0}^{\infty} [1-g(x_0|a)]\varrho_R(a|a_0)da.$$

Though these expressions arise in a purely mathematical way, they can be interpreted physically. The expression $k_{0R}(x_0|a_0)dx_0$, e.g., gives the probability that an isolated particle that starts at a random position in accordance with the density $\varrho_R(a|a_0)$ occupies, at time t, a position between x_0 and x_0+dx_0. Analogously, the expression $k_{LR}(x_0|a_0)$ coincides with the probability that this particle is to be found at an arbitrary position to the left of x_0, i.e., it started as a right neighbor but would now, if the particle-particle interaction were absent, be on the left-hand side of the tagged particle.

All the quantities defined in Eq. (42) do not depend anymore on the particular values of the indices i and j, but only on their signs, i.e., only on the *types* of factor and operation. This implies that all terms $\overline{T}_\pi(x_0|a_0)$ that contain the same *numbers* of these quantities are equal. It is now combinatorics that tells us what patterns of products out of these quantities are possible and how many (identical) terms $\overline{T}_\pi(x_0|a_0)$ belong to each of these patterns.

All in all, there are $(R_L+1+R_R)!$ permutations of the R_L+1+R_R coordinates and, consequently, as many terms $\overline{T}_\pi(x_0|a_0)$ in the sum of $p(x_0|a_0)$. At first, we consider the type of factor of $T(x_0|\mathbf{V}_\pi\mathbf{a})$ that remains without integration.

Case 0. The 0-type operation is applied to the 0-type factor. Consequently, the resulting term $\overline{T}_\pi(x_0|a_0)$ contains k_{00}. All other factors of this term can be either k_{LL}, k_{LR}, k_{RL}, or k_{RR}. Their respective numbers are h_{LL}, h_{LR}, h_{RL}, and h_{RR}. The task to be solved is to determine how many possibilities there are to allot $r_L^o = R_L$ L-type operations and $r_R^o = R_R$ R-type operations on $r_L^f = R_L$ L-type factors and $r_R^f = R_R$ R-type factors in such a way that h_{LL} factors of the kind k_{LL}, h_{LR} of the kind k_{LR}, etc., are formed. At first, one observes that

$$h_{LL}+h_{LR} = r_L^f, \quad h_{RL}+h_{RR} = r_R^f, \quad (43)$$

$$h_{LL}+h_{RL} = r_L^o, \quad h_{LR}+h_{RR} = r_R^o.$$

This means that if, e.g., h_{LL} is chosen, all other numbers are determined:

$$h_{LL}=h, \quad h_{LR}=r_L^f-h, \quad h_{RL}=r_L^o-h, \quad h_{RR}=h+r_R^o-r_L^f. \quad (44)$$

The following list gives the successive choices that have to be made in order to select a special combination of operations and factors, together with the corresponding number of possibilities if h_{LL} shall attain a given value h.

(i) $\binom{r_L^o}{h}$: Out of the L-type operations, $h_{LL}=h$ are chosen to be applied to L-type factors. (Consequently, the remaining L-type operations are applied to R-type factors.)

(ii) $\binom{r_B^0}{r_L^f-h}$: Out of the R-type operations, $h_{LR}=r_L^f-h$ are chosen to be applied to the remaining L-type factors. (Consequently, the remaining R-type operations are applied to the remaining R-type factors.)

(iii) $r_L^f!$: The operations that have been chosen for the L-type factors can be arranged into an arbitrary order (i.e., permutated).

(iv) $r_R^f!$: Likewise, the operations that have been chosen for the R-type factors can be arranged.

The number of (equal) terms with $h_{LL}=h$ that fall into case 0 is the product of these numbers; if the values of r_L^f, etc., of case 0 are inserted one gets

$$H^0(h)=\binom{R_L}{h}\binom{R_R}{R_L-h}R_L!R_R!. \tag{45}$$

The range of allowed values h depends on R_L and R_R. Here and in the following we restrict ourselves, without loss of generality, to the case $R_L\leqslant R_R$ and make use of the abbreviation

$$\Delta R=R_R-R_L\geqslant 0. \tag{46}$$

Then the variable $h=h_{LL}$ can take all values from 0 to R_L. The total number of case-0 terms is $(R_L+1+R_R)!/(R_L+1+R_R)=(R_L+R_R)!$ because we have chosen the 0-type factor $f(x_0|a_0)$ out of all the R_L+1+R_R factors. This can be used to test Eq. (45): The validity of the identity $\Sigma_{h=0}^{R_L}H^0(h)=(R_L+R_R)!$ confirms the result. Now we are able to write down the sum of all terms that belong to case 0:

$$S^0=\sum_{h=0}^{R_L}H^0(h)k_{00}k_{LL}^h k_{LR}^{R_L-h}k_{RL}^{R_L-h}k_{RR}^{h+\Delta R}. \tag{47}$$

Case L. The factor that remains without integration is of L type. The resulting term of $p(x_0|a_0)$ therefore contains k_{L0}. This case splits further into subcases according to whether the operation acting on the factor $f(x_0|a_0)$ is of L-type or R-type.

Subcase LL. The resulting term additionally contains the factor k_{0L}. The remaining R_L+R_R-1 factors can, as in case 0, be either k_{LL}, k_{RL}, k_{LR}, or k_{RR}. One now has $r_L^f=R_L-1$ (one of the L-type factors is already ''consumed'' by the 0-type operation), $r_R^f=R_R$, $r_L^o=R_L-1$ (one of the L-type operations is already applied to the 0-type factor), and $r_R^o=R_R$. In addition to the possible choices in case 0 one has, independent of the numbers h_{LL}, etc., R_L possibilities to choose the L-type factor to which the 0-type operation is applied and further R_L possibilities to choose the L-type operation that is applied to the 0-type factor. Thus

$$H^{LL}(h)=\binom{R_L-1}{h}\binom{R_R}{R_L-1-h}(R_L-1)!R_R!R_L^2. \tag{48}$$

The maximal value of $h=h_{LL}$ is reduced to R_L-1. The total number of terms belonging to subcase LL is

$$(R_L+1+R_R)!\frac{R_L}{R_L+1+R_R}\frac{R_L}{R_L+R_R}=(R_L+R_R-1)!R_L^2$$

because of the probabilities of choosing the L-type factor out of all factors and the L-type operation out of the L-type or R-type operations. The sum of all terms of the subcase reads

$$S^{LL}=\sum_{h=0}^{R_L-1}H^{LL}(h)k_{L0}k_{0L}k_{LL}^h k_{LR}^{R_L-1-h}k_{RL}^{R_L-1-h}k_{RR}^{h+\Delta R+1}. \tag{49}$$

Subcase LR. The resulting term additionally contains the factor k_{0R}. One has $r_L^f=R_L-1$, $r_R^f=R_R$, $r_L^o=R_L$, and $r_R^o=R_R-1$ and there are R_L possibilities of choosing the L-type factor and R_R possibilities of choosing the R-type operation, so that

$$H^{LR}(h)=\binom{R_L}{h}\binom{R_R-1}{R_L-1-h}(R_L-1)!R_R!R_LR_R, \tag{50}$$

$$S^{LR}=\sum_{h=0}^{R_L-1}H^{LR}(h)k_{L0}k_{0R}k_{LL}^h k_{LR}^{R_L-1-h}k_{RL}^{R_L-h}k_{RR}^{h+\Delta R}. \tag{51}$$

Case R. Analogous considerations yield the expressions for the subcases RL and RR:

$$H^{RL}(h)=\binom{R_L-1}{h}\binom{R_R}{R_L-h}R_L!(R_R-1)!R_LR_R, \tag{52}$$

$$S^{RL}=\sum_{h=0}^{R_L-1}H^{RL}(h)k_{R0}k_{0L}k_{LL}^h k_{LR}^{R_L-h}k_{RL}^{R_L-1-h}k_{RR}^{h+\Delta R}, \tag{53}$$

$$H^{RR}(h)=\binom{R_L}{h}\binom{R_R-1}{R_L-h}R_L!(R_R-1)!R_R^2, \tag{54}$$

$$S^{RR}=\sum_{h=\left\{\substack{0\\1}\right\}}^{R_L}H^{RR}(h)k_{R0}k_{0R}k_{LL}^h k_{LR}^{R_L-h}k_{RL}^{R_L-h}k_{RR}^{h+\Delta R-1}. \tag{55}$$

In subcase RR, the range of h needs special attention: For $R_L<R_R$, the range comprises, as in case 0, all values from 0 to R_L, but for $R_L=R_R$ there is at least one L-type operation more than R-type factors so that h cannot be less than 1. Thus the summation starts at $h=0$ if $R_L<R_R$ and at $h=1$ if $R_L=R_R$. The summation range in both S^{RR} and S^{RL}, however, can be altered into the full range from 0 to R_L without changing the sum because the added terms contain $\binom{R_L-1}{R_L}=0$.

Now the set of cases is complete and the propagator can be written as

$$p(x_0|a_0)=\frac{1}{Z}(S^0+S^{LL}+S^{LR}+S^{RL}+S^{RR}). \tag{56}$$

If the abbreviation

$$y:=\frac{k_{RR}k_{LL}}{k_{RL}k_{LR}} \tag{57}$$

is introduced, the five terms may be simplified in the following way:

$$\frac{1}{Z}S^{0}=k_{00}k_{LR}^{R_L}k_{RL}^{R_L}k_{RR}^{\Delta R}\sum_{h=0}^{R_L}\binom{R_L}{h}\binom{R_R}{R_L-h}y^{h},$$

$$\frac{1}{Z}S^{LL}=R_L k_{L0}k_{0L}k_{LR}^{R_L-1}k_{RL}^{R_L-1}k_{RR}^{\Delta R+1}\sum_{h=0}^{R_L-1}\binom{R_L-1}{h}\times\binom{R_R}{R_L-1-h}y^{h},$$

$$\frac{1}{Z}S^{LR}=R_R k_{L0}k_{0R}k_{LR}^{R_L-1}k_{RL}^{R_L}k_{RR}^{\Delta R}\sum_{h=0}^{R_L-1}\binom{R_L}{h}\binom{R_R-1}{R_L-1-h}y^{h},$$

$$\frac{1}{Z}S^{RL}=R_L k_{R0}k_{0L}k_{LR}^{R_L}k_{RL}^{R_L-1}k_{RR}^{\Delta R}\sum_{h=0}^{R_L}\binom{R_L-1}{h}\binom{R_R}{R_L-h}y^{h},$$

$$\frac{1}{Z}S^{RR}=R_R k_{R0}k_{0R}k_{LR}^{R_L}k_{RL}^{R_L}k_{RR}^{\Delta R-1}\sum_{h=0}^{R_L}\binom{R_L}{h}\binom{R_R-1}{R_L-h}y^{h}.$$

These sums can be expressed in terms of Jacobi polynomials $P_n^{(\alpha,\beta)}$ by employing their explicit representation [20]

$$\sum_{k=0}^{n}\binom{n+\alpha}{k}\binom{n+\beta}{n-k}y^{k}=(y-1)^{n}P_n^{(\alpha,\beta)}\left(\frac{y+1}{y-1}\right) \quad (58)$$

and their recurrence relations [20]. Moreover, one easily sees from the definition (42) together with Eq. (38) that

$$k_{R0}=1-k_{L0},\quad k_{LL}=1-k_{RL},\quad k_{RR}=1-k_{LR}, \quad (59)$$

whence k_{R0}, k_{LL}, and k_{RR} can be substituted. Using the abbreviation

$$z:=\frac{y+1}{y-1}=\frac{2k_{LR}k_{RL}}{1-k_{LR}-k_{RL}}+1, \quad (60)$$

the final result is

$$\begin{aligned}p(x_0|a_0)=(1-k_{LR}-k_{RL})^{R_L}(1-k_{LR})^{\Delta R}\Bigg(&k_{00}P_{R_L}^{(0,\Delta R)}(z)\\&+R_L k_{L0}k_{0L}\frac{R_L P_{R_L}^{(0,\Delta R)}(z)+R_R P_{R_L-1}^{(0,\Delta R)}(z)}{(R_L+R_R)(1-k_{RL})}\\&+R_R k_{L0}k_{0R}\frac{R_L P_{R_L}^{(0,\Delta R)}(z)-R_L P_{R_L-1}^{(0,\Delta R)}(z)}{(R_L+R_R)k_{LR}}\\&+R_L(1-k_{L0})k_{0L}\frac{R_R P_{R_L}^{(0,\Delta R)}(z)-R_R P_{R_L-1}^{(0,\Delta R)}(z)}{(R_L+R_R)k_{RL}}\\&+R_R(1-k_{L0})k_{0R}\frac{R_R P_{R_L}^{(0,\Delta R)}(z)+R_L P_{R_L-1}^{(0,\Delta R)}(z)}{(R_L+R_R)(1-k_{LR})}\Bigg).\end{aligned} \quad (61)$$

Summarizing, Eq. (61) gives the propagator of a tagged particle in a single-file system under the following conditions. The tagged particle starts at the initial coordinate a_0. It has R_L neighboring particles to the left whose initial coordinates are randomly distributed according to a common probability density $\varrho_L(a_j|a_0)$. Likewise, it has R_R neighbors to the right, initially distributed according to a density $\varrho_R(a_j|a_0)$. The motion of all individual particles is determined by the isolated-particle propagator $f(x|a)$ and by the hard-core interaction between adjacent particles. These quantities represent the input of Eq. (61), entering $p(x_0|a_0)$ through the auxiliary quantities $k_{L0},k_{00},k_{0L},k_{0R},k_{LR},k_{RL}$, and z as defined by Eqs. (42) and (60), respectively.

D. Example: The infinite channel

In order to illustrate and check the general result (61), consider the special case of a homogeneous infinite channel with the isolated-particle propagator $f_i(x|a)$ according to Eq. (5). Without loss of generality, $a_0=0$ can be assumed. The left and right neighbors of the tagged particle initially are uniformly distributed over finite intervals of lengths L_L and L_R, respectively. The initial densities are therefore given by

$$\varrho_L(a|0)=\begin{cases}0, & a<-L_L\\ 1/L_L, & -L_L\leqslant a\leqslant 0,\end{cases}$$

$$\varrho_R(a|0)=\begin{cases}1/L_R, & 0\leqslant a\leqslant L_R\\ 0, & a>L_R.\end{cases} \quad (62)$$

According to these choices, the auxiliary quantities can be calculated due to their definition (42) by introducing Eqs. (5), (6), and (62) and using Eqs. (A4) and (A5):

$$k_{0L}(x_0|0)=\frac{1}{L_L}\int_{-L_L}^{0}f_i(x_0|a)da=\frac{1}{L_L}[\Gamma(x_0+L_L)-\Gamma(x_0)],$$

$$\begin{aligned}k_{RL}(x_0|0)&=\frac{1}{L_L}\int_{-L_L}^{0}[1-g_i(x_0|a)]da\\&=1-\frac{1}{L_L}[\Lambda(x_0+L_L)-\Lambda(x_0)],\end{aligned}$$

$$k_{L0}(x_0|0)=g_i(x_0|0)=\Gamma(x_0), \quad (63)$$

$$k_{00}(x_0|0)=f_i(x_0|0)=\Phi(x_0),$$

$$k_{LR}(x_0|0)=\frac{1}{L_R}\int_{0}^{L_R}g_i(x_0|a)da=\frac{1}{L_R}[\Lambda(x_0)-\Lambda(x_0-L_R)],$$

$$k_{0R}(x_0|0)=\frac{1}{L_R}\int_{0}^{L_R}f_i(x_0|a)da=\frac{1}{L_R}[\Gamma(x_0)-\Gamma(x_0-L_R)].$$

[The explicit forms of the analytical functions Φ, Γ, and Λ are given by Eqs. (A1), (A2), and (A3), respectively.] Introducing these expressions into Eq. (61) yields the resulting propagator $p_i(x_0|0)$.

An example of this propagator for special values of the parameters R_L, L_L, R_R, and L_R is presented in Fig. 1. The analytical curve due to Eqs. (61) and (63) is compared with the result of a Monte Carlo computer simulation. Like in all examples of this paper, the simulations were carried out on a lattice with ten points per unit length. Considering that point-like particles have to be simulated, neighboring particles

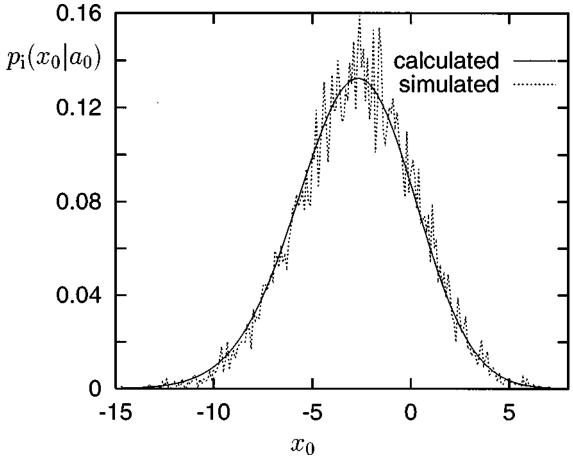

FIG. 1. Example: propagator $p_i(x_0|a_0)$ of a tagged particle in an infinite single file at time $4Dt=100$. The tagged particle starts at $a_0=0$, surrounded by $R_L=2$ left neighbors (initially distributed over the interval $[-2,0]$) and $R_R=8$ right neighbors (initially distributed over the interval $[0,8]$). As expected, there is a drift to the left because at this side there are fewer particles plugging up the way of the tagged particle.

were allowed to occupy the same lattice site but, of course, they could never change their order. The simulated propagators represent relative occurrences based on an ensemble of 10 000 independent identical systems. The coincidence of the simulated and the calculated propagators was checked successfully in the standard way by a χ^2 test of goodness of fit with the significance level $\alpha=0.01$.

Similar calculations can be done on the basis of the propagators $f_{rr}(x|a)$ [Eq. (10)] or $f_{aa}(x|a)$ [Eq. (13)] for finite channels with reflecting or absorbing boundaries, respectively. Although, in these cases, the reflection principle is used twice (first in accounting for the boundaries and second in accounting for the single-file confinement), there is no disturbing interference between both kinds of reflection planes in the state space S. This follows from the general validity of the presented calculation, but may also be realized explicitly by geometrical arguments.

E. Symmetric systems

All special channels considered in Sec. II possess a symmetry point at a certain position S where the propagator can be reflected,

$$f(2S-x|2S-a)=f(x|a). \tag{64}$$

(For the finite channels, this symmetry point lies in the center $S=L/2$, while for the infinite homogeneous channel S can be set at any position.) If the initial distribution of the particles is symmetric as well,

$$\varrho_L(2S-a|2S-a_0)=\varrho_R(a|a_0), \tag{65}$$

the calculation can be simplified using the relations

$$k_{0L}(x_0|a_0)=k_{0R}(2S-x_0|2S-a_0), \tag{66}$$

$$k_{RL}(x_0|a_0)=k_{LR}(2S-x_0|2S-a_0),$$

which follow from the definitions (42) using the identity $g(2S-x|2S-a)=1-g(x|a)$ according to Eqs. (2) and (64). In this case, therefore, it suffices to calculate the quantities k_{00},k_{L0},k_{0R}, and k_{LR}. Note that this is true even if $R_L\neq R_R$.

IV. THE SINGLE-FILE SYSTEM WITH INFINITELY MANY PARTICLES

A. Fourth step: The limit

So far, a system containing a given *finite* number of particles was considered. In many applications, including the examples indicated in the Introduction, however, the number of particles is infinite. The strategy to obtain the propagator in these cases is first to consider an auxiliary system with a finite number of particles and then to take the limit of the propagator if the number of particles tends to infinity. Of course, the auxiliary system has to be designed in such a way that the quantities $R_L\varrho_L(a_j|a_0)$ and $R_R\varrho_R(a_j|a_0)$ tend, in this limit, to the given initial particle concentration of the considered system. This has to be ensured by a suitable dependence of the initial distributions $\varrho_L(a_j|a_0)$ and $\varrho_R(a_j|a_0)$ on the number of particles. Moreover, in some applications also the isolated-particle propagator $f(x|a)$ has to depend in a suitable way on the number of particles in order to prevent the final concentration (after infinite time) from diverging. The general calculation in the present section assumes that this is fulfilled.

In the limit of an infinite number of particles, any (finite) difference ΔR between the numbers of the left and the right neighbors becomes irrelevant. Thus one can set

$$R_L=R_R=:R. \tag{67}$$

The propagator (61) then simplifies to

$$\begin{aligned}p(x_0|a_0)=(1-k_{LR}-k_{RL})^R\Bigg(&k_{00}P_R(z)\\&+k_{L0}Rk_{0L}\frac{P_R(z)+P_{R-1}(z)}{2(1-k_{RL})}\\&+k_{L0}Rk_{0R}\frac{P_R(z)-P_{R-1}(z)}{2k_{LR}}\\&+(1-k_{L0})Rk_{0L}\frac{P_R(z)-P_{R-1}(z)}{2k_{RL}}\\&+(1-k_{L0})Rk_{0R}\frac{P_R(z)+P_{R-1}(z)}{2(1-k_{LR})}\Bigg),\end{aligned} \tag{68}$$

where the Jacobi polynomials have become Legendre polynomials P_R. In order to take the limit of this expression we define quantities m as the limits of the auxiliary quantities k if the number of particles tends to infinity,

$$m_{L0}:=\lim_{R\to\infty}k_{L0}, \quad m_{00}:=\lim_{R\to\infty}k_{00}, \tag{69}$$

and quantities q as the limits of Rk,

$$q_{0L} := \lim_{R\to\infty} Rk_{0L}\,, \quad q_{0R} := \lim_{R\to\infty} Rk_{0R}\,,$$

$$q_{LR} := \lim_{R\to\infty} Rk_{LR}\,, \quad q_{RL} := \lim_{R\to\infty} Rk_{RL}\,, \tag{70}$$

and assume that all these limits exist. The existence of q_{LR} and q_{RL} implies

$$\lim_{R\to\infty} k_{LR}=0, \quad \lim_{R\to\infty} k_{RL}=0. \tag{71}$$

For an arbitrary R-dependent quantity $k=k(R)$ one has

$$\lim_{R\to\infty} (1+k)^R = \exp(q) \quad \text{with} \quad q=\lim_{R\to\infty} Rk \tag{72}$$

if the limit q exists. In this way, one obtains the limit of the first part of the expression (68),

$$\lim_{R\to\infty} (1-k_{LR}-k_{RL})^R = \exp(-q_{LR}-q_{RL}). \tag{73}$$

The limit of the Legendre polynomial can be found via the integral representation [21]

$$P_R(z)=\frac{1}{\pi}\int_0^{\pi}(z\pm\sqrt{z^2-1}\cos\varphi)^R d\varphi. \tag{74}$$

Inserting Eq. (60), the integrand becomes

$$(z\pm\sqrt{z^2-1}\cos\varphi)^R=\left(1+\frac{2k_{LR}k_{RL}\pm 2\sqrt{(1-k_{LR})(1-k_{RL})k_{LR}k_{RL}}\cos\varphi}{1-k_{LR}-k_{RL}}\right)^R. \tag{75}$$

In order to get the limit of this expression one computes, according to Eq. (72),

$$\lim_{R\to\infty} R\,\frac{2k_{LR}k_{RL}\pm 2\sqrt{(1-k_{LR})(1-k_{RL})k_{LR}k_{RL}}\cos\varphi}{1-k_{LR}-k_{RL}} = 2\sqrt{q_{LR}q_{RL}}\cos\varphi,$$

where Eq. (71) was used, and obtains

$$\lim_{R\to\infty} P_R(z)=\frac{1}{\pi}\int_0^{\pi} e^{2\sqrt{q_{LR}q_{RL}}\cos\varphi}d\varphi. \tag{76}$$

This coincides with the integral representation of the modified Bessel functions I_n [20]:

$$I_n(x)=\frac{1}{\pi}\int_0^{\pi} e^{x\cos\varphi}\cos(n\varphi)d\varphi. \tag{77}$$

The result therefore reads

$$\lim_{R\to\infty} P_R(z)=\lim_{R\to\infty} P_{R-1}(z)=I_0(2\sqrt{q_{LR}q_{RL}}). \tag{78}$$

A final difficulty arises from the fact that the fractions in lines 3 and 4 of Eq. (68) yield, in the limit, the indefinite expression 0/0 [cf. Eqs. (71) and (78)]. Using the integral representation (74) again one obtains in line 4

$$\frac{P_R(z)-P_{R-1}(z)}{2k_{RL}}=\frac{1}{\pi}\int_0^{\pi}\frac{(z\pm\sqrt{z^2-1}\cos\varphi)^R}{z\pm\sqrt{z^2-1}\cos\varphi}\,\frac{(z\pm\sqrt{z^2-1}\cos\varphi)-1}{2k_{RL}}d\varphi. \tag{79}$$

The limit of the second fraction of the integrand becomes

$$\lim_{R\to\infty}\frac{(z\pm\sqrt{z^2-1}\cos\varphi)-1}{2k_{RL}}=\lim_{R\to\infty}\frac{k_{LR}+\sqrt{(1-k_{LR})(1-k_{RL})\frac{k_{LR}}{k_{RL}}}\cos\varphi}{1-k_{LR}-k_{RL}}=\sqrt{\frac{q_{LR}}{q_{RL}}}\cos\varphi, \tag{80}$$

while the limit of the denominator of the first fraction, due to Eq. (75), becomes 1. As before, the result turns out to be

$$\lim_{R\to\infty}\frac{P_R(z)-P_{R-1}(z)}{2k_{RL}}=\sqrt{\frac{q_{LR}}{q_{RL}}}\,\frac{1}{\pi}\int_0^{\pi}e^{2\sqrt{q_{LR}q_{RL}}\cos\varphi}\cos\varphi\,d\varphi=\sqrt{\frac{q_{LR}}{q_{RL}}}I_1(2\sqrt{q_{LR}q_{RL}}). \tag{81}$$

The analogous calculation has to be done in line 3. Inserting Eqs. (73), (78), and (81) into Eq. (68) yields the final result

$$\lim_{R\to\infty}p(x_0|a_0)=\exp(-q_{LR}-q_{RL})\Bigg[[m_{00}+m_{L0}q_{0L}+(1-m_{L0})q_{0R}]I_0(2\sqrt{q_{LR}q_{RL}})+\left(m_{L0}q_{0R}\sqrt{\frac{q_{RL}}{q_{LR}}}+(1-m_{L0})q_{0L}\sqrt{\frac{q_{LR}}{q_{RL}}}\right)I_1(2\sqrt{q_{LR}q_{RL}})\Bigg]. \tag{82}$$

Summarizing, Eq. (82) gives the propagator of a tagged particle in the single-file system, surrounded by infinitely many neighboring particles. In order to obtain this propagator, one has to consider an auxiliary system with a finite number of particles and take the limit as the number of particles tend to infinity. If the limits in Eqs. (69) and (70) exist the particle density is sure to remain finite in the limit and the limiting propagator exists.

For symmetric systems obeying Eqs. (64) and (65) the calculation simplifies, as in the case of finitely many particles, according to

$$q_{0L}(x_0|a_0)=q_{0R}(2S-x_0|2S-a_0),$$

$$q_{RL}(x_0|a_0)=q_{LR}(2S-x_0|2S-a_0), \tag{83}$$

whence only the quantities m_{00}, m_{L0}, q_{0R}, and q_{LR} are required.

B. Example: The infinite channel

As announced in the Introduction, the result (82) shall be applied to the infinite, homogeneous, uniformly occupied single file (example 1). As the auxiliary system one can use the system considered in Sec. III D: The infinite channel with the isolated-particle propagator $f_i(x|a)$ due to Eq. (5) where the particles initially are uniformly distributed within finite intervals to the left- or right-hand side of the initial position $a_0=0$ of the tagged particle due to Eq. (62). If one sets

$$L_L=L_R=R/c, \tag{84}$$

with an R-independent constant c, this auxiliary system becomes, in the limit $R\to\infty$, infinite with a homogeneous concentration of c particles per unit length.

Obviously, this system fulfills the symmetry relations (64) and (65) with $S=0$. In order to get the propagator of the tagged particle in this infinite system, according to Sec. IV A one has to calculate the quantities m_{00}, m_{L0}, q_{0R}, and q_{LR} for the special choices of $f(x|a)$, $\varrho_L(a|a_0)$, and $\varrho_R(a|a_0)$ in the present case, which is done using Eqs. (63), (84), and (A9):

$$m_{00}=\lim_{R\to\infty}k_{00}=\Phi(x_0),$$

$$m_{L0}=\lim_{R\to\infty}k_{L0}=\Gamma(x_0),$$

$$q_{0R}=\lim_{R\to\infty}Rk_{0R}=c\Gamma(x_0),$$

$$q_{LR}=\lim_{R\to\infty}Rk_{LR}=c\Lambda(x_0).$$

These expressions are inserted into Eqs. (82) and (83). Using Eq. (A7), the final result reads

$$p_I(x_0|0)=\exp\{-c[\Lambda(x_0)+\Lambda(-x_0)]\}\Bigg[[\Phi(x_0)+2c\Gamma(x_0)\Gamma(-x_0)]I_0[2c\sqrt{\Lambda(x_0)\Lambda(-x_0)}]+c\left(\Gamma^2(x_0)\sqrt{\frac{\Lambda(-x_0)}{\Lambda(x_0)}}+\Gamma^2(-x_0)\times\sqrt{\frac{\Lambda(x_0)}{\Lambda(-x_0)}}\right)I_1[2c\sqrt{\Lambda(x_0)\Lambda(-x_0)}]\Bigg]. \tag{85}$$

Let us consider the asymptotic behavior of the propagator if the mean distance that an isolated particle would have moved is still very small or already very large in comparison to the mean distance between adjacent particles. In the first case $\sqrt{4Dt}\ll 1/c$, the isolated particle has most probably not yet ''felt'' any influence by neighboring particles. This situation occurs for a very small observation time t, a very low diffusivity D, or a very low particle concentration c. It will be referred to as a short-time limit (though it could, of course, equally be termed a low-diffusivity limit or low-concentration limit). In the opposite case $\sqrt{4Dt}\gg 1/c$, the long-time limit, the interactions of the tagged particle with its neighbors predominate its propagation. This should occur after a long observation time, at very rapid diffusion, or in a very crowded channel. In addition to the asymptotic form of the propagator, we are particularly interested in the mean-square displacement of the tagged particle,

$$\langle x_0^2\rangle=\int_{-\infty}^{\infty}x_0^2p_I(x_0|0)dx_0, \tag{86}$$

because of its central relevance for the observation of diffusion phenomena [10,22].

The discussion can be facilitated if the mean distance between adjacent particles $1/c$ is taken as the unit of the length scale: The coordinate x_0 is replaced by the scaled displacement $\tilde{x}_0 = cx_0$ and the diffusion coefficient D by $\tilde{D} = c^2 D$, while the concentration parameter vanishes, $\tilde{c} = 1$. Thus the case of an arbitrary concentration can be mapped onto the case $c = 1$. This implies that one may assume $c \approx 1$ in all order-of-magnitude estimations.

In the short-lime limit, the tagged particle is expected to behave like an isolated particle. To check this, take, without loss of generality, $x_0 \geqslant 0$. For $4Dt \ll 1$ one has, according to Eqs. (A2) and (A3), $\Gamma(x_0) \approx 1$, $\Gamma(-x_0) \approx 0$, $\Lambda(x_0) \approx x_0$, $\Lambda(-x_0) \approx 0$, $I_0 \approx 1$, and $I_1 \approx 0$ and the propagator becomes

$$p_I(x_0|0) \approx \exp(-c|x_0|)\Phi(x_0) \approx \Phi(x_0) = f_i(x_0|0),$$

where the second approximation follows from $c|x_0| \ll x_0^2/4Dt$ in the exponent. Thus the propagator of the tagged particle can indeed be approximated by the propagator of the free particle, which is a Gaussian with the mean-square displacement

$$\langle x_0^2 \rangle = 2Dt. \tag{87}$$

In order to obtain an approximation of the propagator in the opposite case, the long-lime limit, write Eq. (85) in an alternative way by splitting the exponential term into two parts and attaching one of them to the Bessel functions:

$$p_I(x_0|0) = \exp\{-c[\sqrt{\Lambda(x_0)} - \sqrt{\Lambda(-x_0)}]^2\} c \left[\left(\frac{\Phi(x_0)}{c} + 2\Gamma(x_0)\Gamma(-x_0) \right) e^{-\lambda} I_0(\lambda) \left(\Gamma^2(x_0) \frac{\sqrt{\Lambda(-x_0)}}{\sqrt{\Lambda(x_0)}} + \Gamma^2(-x_0) \frac{\sqrt{\Lambda(x_0)}}{\sqrt{\Lambda(-x_0)}} \right) e^{-\lambda} I_1(\lambda) \right], \tag{88}$$

$$\text{with } \lambda := 2c\sqrt{\Lambda(x_0)\Lambda(-x_0)}.$$

Then investigate the behavior of the functions Φ, Γ, and Λ for $4Dt \gg 1$. For any coordinate x_0 the relation

$$\xi := \frac{x_0}{\sqrt{4Dt}} \ll 1 \tag{89}$$

holds as soon as $4Dt$ becomes sufficiently large. Then the functions can be expanded,

$$\Phi(\pm x_0) = \frac{1}{\sqrt{\pi}\sqrt{4Dt}}[1 + O(\xi^2)], \tag{90}$$

$$\Gamma(\pm x_0) = \frac{1}{2}\left(1 \pm \frac{2}{\sqrt{\pi}}\xi + O(\xi^2)\right), \tag{91}$$

$$\Lambda(\pm x_0) = \frac{\sqrt{4Dt}}{2\sqrt{\pi}}[1 \pm \sqrt{\pi}\xi + O(\xi^2)], \tag{92}$$

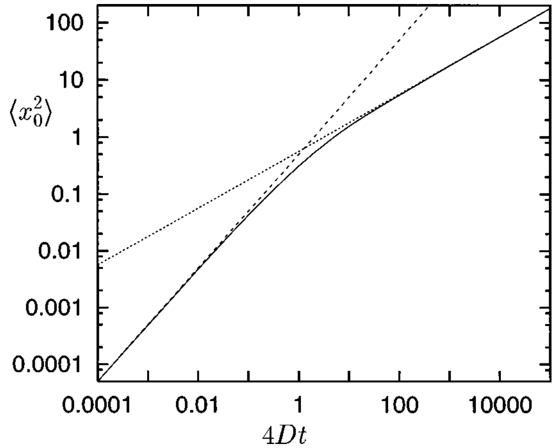

FIG. 2. Mean-square displacement of a tagged particle in an infinite single file, homogeneously occupied with $c = 1$ particles per unit length. The broken lines $2Dt$ and $\sqrt{4Dt}/\sqrt{\pi}$, respectively, indicate the asymptotic behavior.

$$\sqrt{\Lambda(\pm x_0)} = \sqrt{\frac{\sqrt{4Dt}}{2\sqrt{\pi}}}\left(1 \pm \frac{\sqrt{\pi}}{2}\xi + O(\xi^2)\right). \tag{93}$$

Turning to the large square brackets of Eq. (88), one observes that all terms $\Gamma(\pm x_0)$ become approximately 1/2. Compared with this, the term $\Phi(x_0)$ can be canceled due to $\sqrt{4Dt}$ in the denominator. The fractions $\sqrt{\Lambda(\pm x_0)}/\sqrt{\Lambda(\mp x_0)}$ tend to 1 and therefore vanish as well. The only surviving contributions are the Bessel functions, which can be approximated according to $e^{-\lambda} I_n(\lambda) \approx 1/\sqrt{2\pi\lambda}$, valid for large λ, because λ increases with t. If the expansion of $\sqrt{\Lambda(\pm x_0)}$ due to Eq. (93) is inserted into λ and into the exponential factor of Eq. (88) one arrives at

$$p_I(x_0|0) \approx \frac{1}{\sqrt{2\pi \frac{\sqrt{4Dt}}{c\sqrt{\pi}}}} \exp\left(-\frac{x_0^2}{2\frac{\sqrt{4Dt}}{c\sqrt{\pi}}} \right), \tag{94}$$

which is nothing but a Gaussian with the mean-square displacement

$$\langle x_0^2 \rangle = \frac{1}{\sqrt{\pi}c}\sqrt{4Dt}. \tag{95}$$

This finding coincides, as expected, with the asymptotic behavior given in [7] for the propagator derived there.

Thus we have confirmed that both for small and for large observation times the propagator of the tagged particle in the infinite single-file channel tends to a Gaussian with a mean-square displacement according to Eq. (87) or (95), respectively. However, Eq. (85) is valid at intermediate observation times as well. Figure 2 shows the mean-square displacement, obtained by numerical integration according to Eqs. (85) and (86), as a function of the scaled observation time. The figure clearly shows the transition between the two limiting regimes of propagation, which are indicated by the broken lines. [If the calculation were based on the telegrapher's equation (7) rather than the diffusion equation (3), there

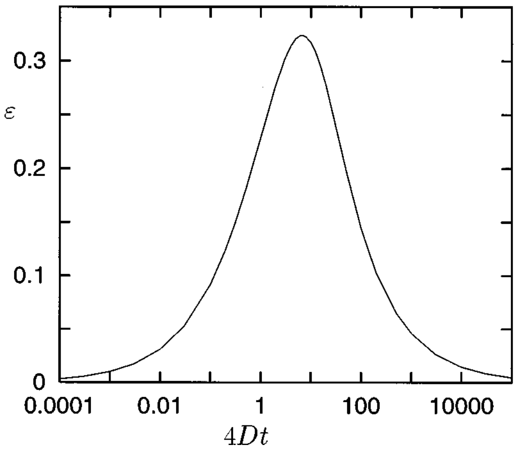

FIG. 3. Excess ε of the propagator $p_I(x_0|0)$ with $c=1$ as a function of the observation time, expressing the deviation of the propagator from a Gaussian of identical variance.

would be two superimposing transitions: first, from the ballistic to the diffusive behavior of the isolated particles and, second, as described, from the free to the single-file behavior.] In order to assess the deviation of the propagator $p_I(x_0|0)$ from a Gaussian with identical mean-square displacement, one inspects in Fig. 3 its excess

$$\varepsilon=\frac{\langle x_0^4\rangle}{\langle x_0^2\rangle^2}-3 \tag{96}$$

presented over the same range of observation times as in Fig. 2. Obviously, the maximal deviation occurs roughly halfway through the transition. The explicit form of the propagator at this maximum is given in Fig. 4 and compared with the corresponding Gaussian. Obviously, the difference between both curves is rather small. If, therefore, the propagator is fitted to experimental data whose error is larger than this difference, it can, for all observation times, be approximated by a Gaussian.

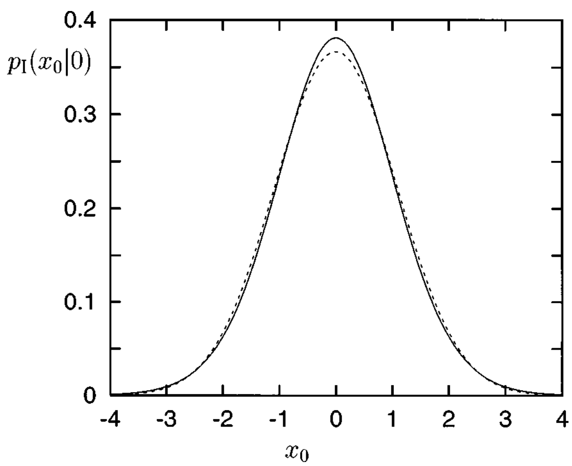

FIG. 4. Example: propagator $p_I(x_0|0)$ of a tagged particle in an infinite single file, homogeneously occupied with $c=1$ particles per unit length, at time $4Dt=6.5$ (solid line), compared with a Gaussian of identical variance (broken line). At this time, the excess ε of the propagator is maximal.

As in Sec. III D, the result (85) was checked by comparison with computer simulations via a χ^2 test. In the simulation, the infinite channel was approximated by 1000 neighboring particles on either side.

C. Example: Absorbing boundaries

Let us now turn to the finite channel with absorbing boundaries (cf. example 2 in the Introduction). Though the number of particles within this finite channel is, of course, finite, the initial equilibration with the infinite particle reservoir involves an infinite number of particles. Moreover, since the particle concentration c (rather than the number of particles) within the channel is given, the actual number is allowed to fluctuate around the average value cL. This can only be accounted for by considering an infinite number of particles. Thus this example demonstrates that the limit of infinitely many particles can be essential even for finite single-file channels.

The system can be modeled as follows. Initially, a small (finite) number of particles is placed into the channel in such a way that it builds up the homogeneous particle concentration c, while the (infinite) rest is placed into the reservoir outside the file. For $t\geqslant 0$ when the desorption process has started, the individual particles behave according to the isolated-particle propagator $f_{aa}(x|a)$ considered in Sec. II D: As soon as a particle reaches one of the boundaries, it is swallowed by the vacuum and will never return.

In the auxiliary system, where the number of particles is finite, we adjust the initial densities in such a way that the particle concentration within the file has its given value c, while the (still finite) rest of the particles are placed in the reservoirs:

$$\varrho_L(a|a_0)=\begin{cases} \ldots, & 0<a \\ c/R, & 0\leqslant a\leqslant a_0\end{cases}$$

$$\varrho_R(a|a_0)=\begin{cases} c/R, & a_0\leqslant a\leqslant L \\ \ldots, & L<a\end{cases}; \tag{97}$$

the ellipses stand for arbitrary distributions outside the channel ensuring Eq. (38), which will not enter into the result. Again, the system is symmetrical, where $S=L/2$. With $f_{aa}(x|a)$ due to Eq. (13), $g_{aa}(x|a)$ due to Eq. (15), and these initial densities one has (for $0\leqslant x_0\leqslant L$)

$$k_{00}(x_0|a_0)=f_{aa}(x_0|a_0),$$

$$k_{L0}(x_0|a_0)=g_{aa}(x_0|a_0),$$

$$k_{0R}(x_0|a_0)=\frac{c}{R}\int_{a_0}^{L}f_{aa}(x_0|a)da+\int_L^\infty f_{aa}(x_0|a)\cdots da, \tag{98}$$

$$k_{LR}(x_0|a_0)=\frac{c}{R}\int_{a_0}^{L}g_{aa}(x_0|a)da+\int_L^\infty g_{aa}(x_0|a)\cdots da.$$

The ellipses are the arbitrary distribution from Eq. (97). The terms containing them vanish because $f_{aa}(x_0|a)=0$ and $g_{aa}(x_0|a)=0$ for $a>L$ according to Eqs. (13) and (15).

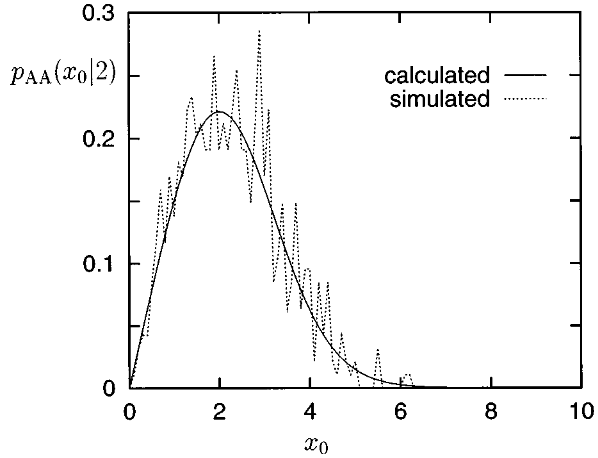

FIG. 5. Example: propagator $p_{AA}(x_0|a_0)$ of a tagged particle in a finite single file of length $L=10$ with two absorbing boundaries, initially homogeneously occupied with $c=1$ particles per unit length, at time $4Dt=10$. The tagged particle starts at $a_0=2$.

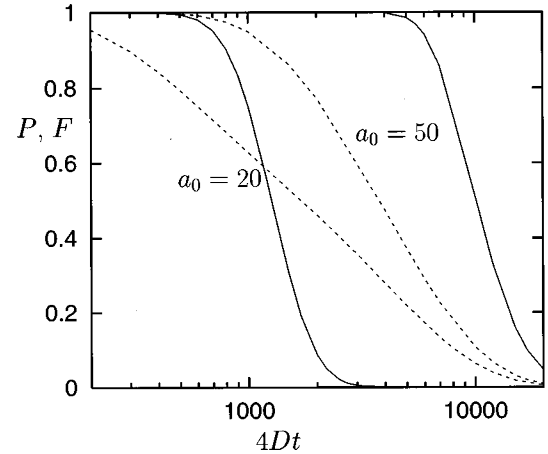

FIG. 6. Probability that a given particle is still inside a finite channel of length $L=100$ with absorbing boundaries. Initially, the channel is occupied with $c=1$ particles per unit length. The tagged particle is identified according to its starting position (here $a_0=50$ or $a_0=20$, respectively). The situation in single-file systems (solid lines) is compared with the case of noninteracting particles (broken lines).

Now the limit $R\rightarrow\infty$ is performed. With Eqs. (69) and (70) one calculates

$$m_{00}(x_0|a_0)=f_{aa}(x_0|a_0),$$

$$m_{L0}(x_0|a_0)=g_{aa}(x_0|a_0),$$

$$q_{0R}(x_0|a_0)=c\int_{a_0}^{L}f_{aa}(x_0|a)da, \tag{99}$$

$$q_{LR}(x_0|a_0)=c\int_{a_0}^{L}g_{aa}(x_0|a)da,$$

where the integrals are given by $(0\leqslant x_0\leqslant L)$

$$\int_{a_0}^{L}f_{aa}(x_0|a)da=\sum_{k=-\infty}^{\infty}[\Gamma(x_0-a_0-2kL)-\Gamma(x_0-L-2kL)+\Gamma(x_0+a_0-2kL)-\Gamma(x_0+L-2kL)], \tag{100}$$

$$\int_{a_0}^{L}g_{aa}(x_0|a)da=\sum_{k=0}^{\infty}[\Lambda(-x_0+a_0+2kL)+\Lambda(x_0+a_0+2kL)+\Lambda(-x_0+2L-a_0+2kL)+\Lambda(x_0+2L-a_0+2kL)-2\Lambda(-x_0+L+2kL)-2\Lambda(x_0+L+2kL)]. \tag{101}$$

If this is introduced into Eq. (82) one obtains the searched propagator $p_{AA}(x_0|a_0)$. As in Sec. III D, an example is given graphically in Fig. 5 and compared with computer simulations. Once more, the χ^2 test confirms the coincidence.

The propagator can be used to calculate the probability that a particular particle is, at a given time, still inside the channel. Assuming that the considered particle starts at a position a_0, this probability is given by

$$P(a_0,t)=\int_0^L p_{AA}(x_0|a_0)dx_0. \tag{102}$$

For the special value $L=100$, $a_0=50$ (the particle starts in the channel center), it is shown in Fig. 6 as a function of time (rightmost solid curve). For comparison, the corresponding probability $F(a_0,t)$ for a free particle is given (broken curve), which can be calculated via an integral similar to Eq. (102) using the isolated-particle propagator $f_{aa}(x_0|a_0)$ rather than the single-file propagator $p_{AA}(x_0|a_0)$. As expected, the free particle leaves, on average, the channel much earlier than the particle subject to single-file confinement. Interestingly enough, also the opposite situation occurs, as is shown by the left two curves in Fig. 6. In this case, the particles start at a position closer to the boundary, $a_0=20$. While the unrestricted particle is free to move towards the channel center where it is far from the absorbing boundary, the single-file particle is not able to leave the vicinity of the boundary whence it eventually has a greater chance to be absorbed. This is illustrated in more detail by Fig. 7, where the probabilities $P(a_0,t)$ and $F(a_0,t)$ for the single-file or the free particle, respectively, are compared at a fixed time dependent on their starting position a_0. The crossover of the two curves is, however, not surprising: As already mentioned in the Introduction, the total mass transport is not influenced by the single-file confinement, so that the mean number of remaining particles within the channel is insensitive to whether or not the particles are able to change their order. This implies that the integrals over both profiles of Fig. 7 are equal,

$$c\int_0^L P(a_0,t)da_0=c\int_0^L F(a_0,t)da_0. \tag{103}$$

The quantitative result presented in Fig. 7 shows that the release of particles from a single-file channel depends much more pronouncedly on their initial positions than in the case of noninteracting particles. This is a characteristic feature of

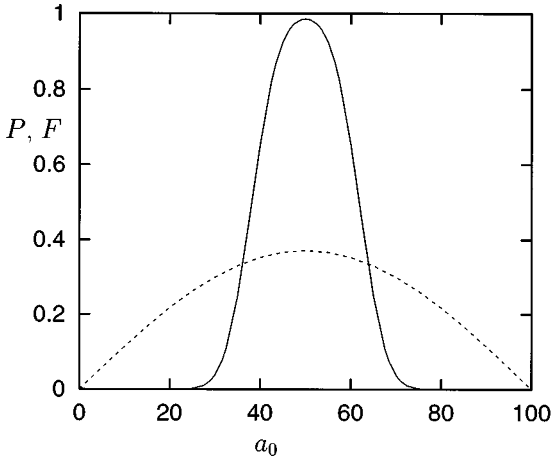

FIG. 7. Probability that a given particle is, at time $4Dt=5000$, still inside a finite channel of length $L=100$ with absorbing boundaries, dependent on its starting position a_0. If starting in the channel center, the particle subject to single-file confinement (solid line) is much less probably absorbed than the free particle (broken line), while the contrary situation is true for particles starting near the boundaries. Initially, the channel is occupied with $c=1$ particles per unit length.

single-file systems which could, e.g., be employed for the sequential release of several particle species if there is an initial spatial order of the species inside the channel.

V. CONCLUSION

The presented formalism yields exact expressions for the propagators of tagged particles in single-file systems. It is valid for arbitrary interactions between the particles and the channel, as described by the isolated-particle propagator $f(x|a)$, and for arbitrary initial distributions of the particles, as described by the probability densities $\varrho_L(a|a_0)$ and $\varrho_R(a|a_0)$. The number of particles may be finite [Eq. (61)] or infinite [Eq. (82)]. As examples, the infinite channel and the finite channel with two absorbing boundaries were considered. The validity of all explicit results was checked by comparison with propagators from computer simulations. As a further check for the homogeneously occupied infinite channel, the asymptotic behavior was compared with results known from the literature.

In addition to the derivation of general expressions of the propagators, practical conclusions were drawn from the results. First, it was shown that the propagator of a tagged particle in an infinite, homogeneously occupied single file deviates, for all observation times, only slightly from a Gaussian. This justifies an assumption often made on evaluating scattering experiments. Second, the release of particles from a finite single file can be investigated quantitatively. One finds a characteristic, strong dependence on the initial positions of the particles within the channel. Particles from the channel center are released much more slowly than in systems ruled by normal diffusion with equal length and diffusivity.

The basic idea of the approach presented is to solve the diffusion equation (or any equivalent differential equation) in the state space S of all particles, where the particle-particle interaction is described by appropriate boundary conditions (or, even more generally, by a total interaction potential in S representing the sum of all pairwise particle-particle interaction potentials). In the case of hard-core repulsion, these boundaries are totally reflecting and can be accounted for by the well-known reflection principle. Currently, we are investigating the generalizations of this method to more complicated systems, including incomplete mutual repulsion of the particles and systems where the single-file behavior is restricted to certain regions of the x axis.

If the particles cannot be considered pointlike, but have a given radius r, the reflection planes in the state space S have to be translated. In this case, the treatment involves additional complications. For some systems, however, where the particle density does not change with time, the propagators may approximately be corrected by an appropriate scaling of the x axis as suggested in [7]: If all the space occupied by particles is cut out, a modified system with again pointlike particles is obtained. This can, on average, be done by scaling the x axis by the factor $1-2rc$ giving the relative amount of unoccupied space.

ACKNOWLEDGMENTS

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APPENDIX

Because of their frequent use in the examples we define the analytical functions

$$\Phi(u):=\frac{1}{\sqrt{4\pi Dt}}\exp\left(-\frac{u^2}{4Dt}\right), \tag{A1}$$

$$\Gamma(x):=\frac{1}{2}\left[1+\mathrm{erf}\left(\frac{x}{\sqrt{4Dt}}\right)\right], \tag{A2}$$

$$\Lambda(x):=\frac{4Dt}{2}\Phi(x)+x\Gamma(x), \tag{A3}$$

with the observation time t and the diffusion coefficient D as parameters. Since these parameters exclusively occur in the form $4Dt$, this expression can be considered as a scaled time, corresponding to twice the mean-square displacement of an isolated particle at this physical time t in a homogeneous channel with the diffusion coefficient D; cf. Eq. (87). The functions are related to each other by the integrals

$$\int_{-\infty}^{x}\Phi(u)du=\Gamma(x), \tag{A4}$$

$$\int_{-\infty}^{x} \Gamma(u) du = \Lambda(x). \tag{A5}$$

Moreover, they fulfill the relations

$$\Phi(-u) = \Phi(u), \tag{A6}$$

$$\Gamma(-x) = 1 - \Gamma(x), \tag{A7}$$

$$\Lambda(-x) = \Lambda(x) - x. \tag{A8}$$

Their limits are

$$\lim_{x \to -\infty} \Phi(x) = 0, \quad \lim_{x \to -\infty} \Gamma(x) = 0, \quad \lim_{x \to -\infty} \Lambda(x) = 0. \tag{A9}$$

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