Active Transport in Dense Diffusive Single-File Systems

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We study a minimal model of active transport in crowded single-file environments which generalizes the emblematic model of single-file diffusion to the case when the tracer particle (TP) performs either an autonomous directed motion or is biased by an external force, while all other particles of the environment (bath) perform unbiased diffusions. We derive explicit expressions, valid in the limit of high density of bath particles, of the full distribution $P^{(n)}(X)$ of the TP position and of all its cumulants, for arbitrary values of the bias f and for any time n. Our analysis reveals striking features, such as the anomalous scaling $\propto \sqrt{n}$ of all cumulants, the equality of cumulants of the same parity characteristic of a Skellam distribution and a convergence to a Gaussian distribution in spite of asymmetric density profiles of bath particles. Altogether, our results provide the full statistics of the TP position and set the basis for a refined analysis of real trajectories of active particles in crowded single-file environments.

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Introduction.—Single-file diffusion refers to one-dimensional diffusion of interacting particles that cannot bypass each other. Clearly, in such a geometry, the initial order of particles remains the same over time, and this very circumstance appears so crucial that the movements of individual particles become strongly correlated: the displacement of any given tracer particle (TP) on progressively larger distances necessitates the motion of more and more other particles in the same direction. This results in a subdiffusive growth of the TP mean-square displacement $\overline{X^2} \sim \sqrt{t}$, first discovered analytically by Harris [1] and subsequently reestablished for systems with differently organized dynamics (see, e.g., Refs. [2–8]). Nowadays, a single-file diffusion, prevalent in many physical, chemical, and biological processes, has been experimentally evidenced by passive microrheology in zeolites, transport of confined colloidal particles, or charged spheres in circular channels [9–13]. It provides a paradigmatic example of anomalous diffusion in crowded *equilibrium* systems, which emerges due to a cooperative many-particle behavior.

On the other hand, systems that consume energy for propulsion—*active* particle systems—have received growing attention in the last decade, both because of the new physical phenomena that they display and their wide range of applications. Examples include self-propelled particles, such as molecular motors or motile living cells [14], and externally driven particles, such as probes in active microrheology experiments [15]. The intrinsic out-of-equilibrium nature of these systems leads to remarkable effects such as non-Boltzmann distributions [16], long-range order even in low spatial dimensions [17], and spontaneous flows [18]. In particular, 1D assemblies of active particles have been extensively studied in the context of asymetric simple exclusion process models (see Ref. [19] for a recent review).

However, up to now, active transport in diffusive single-file systems, which involves an active TP performing an autonomous directed motion or pulled by a constant external force f in a 1D bath of unbiased diffusive particles with hard-core interactions, has drawn uncomparably less attention [20]. Such dynamics, depicted in Fig. 1, provides a minimal model of active transport in crowded single-file environments, which schematically mimics situations as varied as the active transport of a vesicle in a crowded axone [21], directed cellular movements in crowded channels [22], or active microrheology in capillaries [15]. In this context, the only available theoretical results concern the large time behavior of the mean displacement $\bar{X}$ of the TP, which has been shown to grow sublinearly with time $\bar{X} \sim \sqrt{t}$ [23–26]. In fact, the biased TP drives the bath particles to a *nonequilibrium* state with an asymmetric distribution: the bath particles accumulate in front of the TP, thus increasing the frictional force, and are depleted behind. The extent of these perturbations grows in time in proportion to $\sqrt{t}$ and characterizes a subtle interplay between the bias, formation of nonequilibrium density profiles, and backflow effects of the medium on the TP.

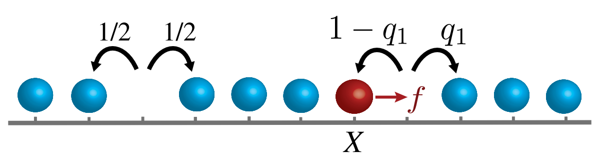

FIG. 1 (color online). Model notations.

In this Letter, we focus on this minimal model of active transport in diffusive single-file systems. Going beyond the previous analysis of the TP mean displacement, we present in the limit of high density of bath particles exact expressions of the full distribution $P^{(n)}(X)$ of the TP position and of all its cumulants for arbitrary values of the bias f and for any time n. In particular, in this high density limit, it is shown that at large times this distribution converges to a Gaussian, with mean $\bar{X} \sim \alpha_f(\rho)\sqrt{n}$ and variance growing asymptotically as $\sqrt{n}$. Remarkably, in this limit, the variance is proved to be independent of f. Altogether, our results provide the full statistics of the TP position and set the basis for a refined analysis of real trajectories of active particles in crowded single-file environments.

The model.—Consider a one-dimensional, infinite in both directions, line of integers x, populated by hard-core particles present at mean density ρ, performing symmetric random walks. At $t = 0$, we introduce at the origin of the lattice an active TP, hopping on its right (respectively, left) neighbor site with probability p_1 (respectively, p_{-1}), which process is also constrained by hard-core exclusion. In what follows, we focus on the limit of a dense system, corresponding to the limit of a small vacancies density $\rho_0 = 1 - \rho \ll 1$. In this limit, it is most convenient to follow the vacancies rather than the particles. We thus formulate directly the dynamics of the vacancies, which unambiguously defines the full dynamics of the system. Following Refs. [27,28], we assume that at each time step, each vacancy is moved to one of its nearest neighbor sites with equal probability. As long as a vacancy is surrounded only by bath particles, it thus performs a symmetrical nearest neighbor random walk. However, due to the biased nature of the movement of the TP, specific rules have to be defined when a vacancy is adjacent to the TP. In this case, if the vacancy occupies the site to the right (respectively, to the left) of the TP, we stipulate that it has a probability $q_1 = 1/(2p_1 + 1)$ [respectively, $q_{-1} = 1/(2p_{-1} + 1)$] to jump to the right (respectively, to the left) and $1 - q_1$ (respectively, $1 - q_{-1}$) to jump to the left (respectively, to the right). These rules are the discrete counterpart of a continuous time version of the model [29], as shown in Ref. [28]. Note that a complete description of the dynamics would require additional rules for cases where two vacancies are adjacent or have common neighbors; however, these cases contribute only to $\mathcal{O}(\rho_0^2)$ and can thus be left unstated.

Single file with a single vacancy.—The dynamics of the TP is controlled by the first-passage statistics of vacancies to the TP position. In the case of an infinitely strong bias $p_1 = 1$, a simple analysis based on this idea (developed later) provides large time asymptotics of the cumulants of the position. However, in the case of a general bias, subtle anticorrelations arise and require the more careful treatment presented below. We start with an auxiliary problem in which the system contains just a single vacancy initially at position Z and which will be proved next to be a key step in the resolution of the complete problem with a (small) concentration of vacancies. Let $p_Z^{(n)}(X)$ denote the probability of having the TP at site X at time moment n, given that the vacancy commenced its random walk at Z. Clearly, in a single-vacancy case, this probability is not equal to zero only for $X = 0$ and $X = 1$, if $Z > 0$, and $X = 0$ and $X = -1$, if $Z < 0$. Following Refs. [27,28], we then represent $p_Z^{(n)}(X)$ as a sum over all passage events of the vacancy to the TP location:

$$p_Z^{(n)}(X) = \delta_{X,\mathbf{0}}\left(1 - \sum_{j=0}^{n} F_Z^{(j)}\right) + \sum_{p=1}^{+\infty} \sum_{m_1,m_2,\ldots,m_p=1}^{+\infty} \times \sum_{m_{p+1}=0}^{+\infty} \delta_{m_1+\cdots+m_{p+1},n}\,\delta_{X,[(\mathrm{sgn}(Z)+(-1)^{p+1})/2]} \times \left(1 - \sum_{j=0}^{m_{p+1}} F_{(-1)^p}^{(j)}\right) F_{(-1)^{p+1}}^{(m_p)} \cdots F_{-1}^{(m_2)} F_Z^{(m_1)}, \quad (1)$$

where $\delta_{a,b} = 1$ when $a = b$ and is equal to zero, otherwise, and $F_Z^{(n)}$ is the probability that the vacancy, which started its random walk at site Z, arrived at the origin for the first time at time moment n. The first term in the right-hand side of Eq. (1) represents the event that at time n, the TP has not been visited by any vacancy, while the second one results from a partition both on the number p of visits and waiting times m_i between visits of the TP by the vacancy.

Now let $\hat{g}(\xi)$ denote the generating function of any time-dependent function $g^{(n)}$, $\hat{g}(\xi) \equiv \sum_{n=0}^{\infty} g^{(n)}\xi^n$. Then, Eq. (1) implies that the generating function of the propagator of the single-vacancy model can be expressed via the generating functions of the corresponding first-passage distributions as

$$\hat{p}_{\pm 1}(X;\xi) = \frac{\delta_{X,0}(1 - \hat{F}_{\pm 1}) + \delta_{X,\pm 1}\hat{F}_{\pm 1}(1 - \hat{F}_{\mp 1})}{(1 - \hat{F}_1\hat{F}_{-1})(1 - \xi)}, \quad (2)$$

where we have used the short notations $\hat{F}_{\pm 1} \equiv \hat{F}_{\pm 1}(\xi)$.

Single file with a small concentration of vacancies.—We now turn to the original problem with a small but finite density ρ_0 of vacancies and aim to express the desired probability $P^{(n)}(X)$ of finding the TP at site X at time n via the propagator for a single-vacancy problem. We consider first a finite chain with L sites, M of which are vacant, and the initial positions of the latter are denoted by Z_j, $j = 1, \ldots, M$. Then, the probability $P^{(n)}(X|\{Z_j\})$ of finding the TP at position X at time moment n as a result of its interaction with all the vacancies collectively, for their fixed initial configuration, writes

$$P^{(n)}(X|\{Z_j\}) = \sum_{Y_1,Y_2,\ldots,Y_M} \delta_{X,Y_1+\cdots+Y_M} P^{(n)}(\{Y_j\}|\{Z_j\}), \quad (3)$$

where $P^{(n)}(\{Y_j\}|\{Z_j\})$ stands for the conditional probability that within the time interval n, the TP has performed a displacement Y_1 due to interactions with the first vacancy, a displacement Y_2 due to the interactions with the second vacancy, etc. In the lowest order in the density of vacancies, the vacancies contribute independently to the total displacement of the tracer, so that the latter conditional probability decomposes

$$P^{(n)}(\{Y_j\}|\{Z_j\}) \sim_{\rho_0\to 0} \prod_{j=1}^{M} p_{Z_j}^{(n)}(Y_j), \tag{4}$$

where $p_{Z_j}^{(n)}(Y_j)$ is the single-vacancy propagator and the symbol $\underset{\rho_0\to 0}{\simeq}$ signifies the leading behavior in the small density of vacancies limit. Note that such an approximation yields results which are exact to the order $\mathcal{O}(\rho_0)$, and hence, such a description is expected to be quite accurate when $\rho_0 \ll 1$ [27,28]. Next, we suppose that initially the vacancies are uniformly distributed on the chain (except for the origin, which is occupied by the TP) and average $P^{(n)}(X|\{Z_j\})$ over the initial distribution of the vacancies. In doing so and subsequently turning to the thermodynamic limit, i.e., setting $L \to \infty$, $M \to \infty$ with $M/L = \rho_0$ kept fixed, we find that the generating function of the second characteristic function

$$\psi_X(k;\xi) \equiv \sum_{n=0}^{\infty} \ln[\tilde{P}^{(n)}(k)]\xi^n, \tag{5}$$

where $\tilde{P}^{(n)}(k) \equiv \sum_{X=-\infty}^{\infty} P^{(n)}(X)e^{ikX}$ satisfies

$$\lim_{\rho_0\to 0}\frac{\psi_X(k;\xi)}{\rho_0} = -\sum_{\epsilon=\pm 1}\left(\frac{1}{1-\xi} - \tilde{p}_{-\epsilon}(k;\xi)e^{i\epsilon k}\right) \times \sum_{Z=1}^{\infty}\hat{F}_{\epsilon Z}(\xi). \tag{6}$$

Our last step consists of the explicit determination of $\hat{F}_{\pm 1}$ and $\sum_{Z=1}^{\infty}\hat{F}_{\epsilon Z}(\xi)$ in Eq. (6). We note that both can be readily expressed via the first-passage time density at the origin at time n of a symmetric one-dimensional Polya random walk, starting at time 0 at position l, denoted as $f_l^{(n)}$, since, by partitioning over the first time when the sites adjacent to the origin are reached, we have

$$F_{\pm 1}^{(n)} = (1-q_{\pm 1})\delta_{n,1} + q_{\pm 1}\sum_{k=1}^{n} f_1^{(k-1)}F_{\pm 1}^{(n-k)}. \tag{7}$$

Multiplying both sides of Eq. (7) by ξ^n, performing summation over n, and taking into account that $\hat{f}_l(\xi) = \sum f_l^{(n)}\xi^n = [(1-\sqrt{1-\xi^2})/\xi]^{|l|}$ [30], we find that

$$\hat{F}_{\pm 1} = \frac{(1-q_{\pm 1})\xi}{1-q_{\pm 1}(1-\sqrt{1-\xi^2})}. \tag{8}$$

Similarly, noticing that

$$F_Z^{(n)} = \begin{cases} \sum_{k=1}^{n} f_{Z-1}^{(k)}F_1^{(n-k)} & \text{if } Z>0 \\ \sum_{k=1}^{n} f_{-1-Z}^{(k)}F_{-1}^{(n-k)} & \text{if } Z<0 \end{cases} \tag{9}$$

and using the definition of $\hat{f}_l(\xi)$ given above, we obtain

$$\sum_{Z=1}^{\infty}\hat{F}_{\pm Z}(\xi) = \frac{\hat{F}_{\pm 1}}{1-(1-\sqrt{1-\xi^2})/\xi}. \tag{10}$$

Gathering the results in Eqs. (7)–(10), substituting them into Eq. (6), we finally derive our central analytical result which defines the exact (in the leading in ρ_0 order) generating function of the cumulants κ_j of arbitrary order j, themselves defined by $\ln[\tilde{P}^{(n)}(k)] \equiv \sum_{j=1}^{\infty}(\kappa_j^{(n)}/j!)(ik)^j$:

$$\lim_{\rho_0\to 0}\frac{\hat{\kappa}_j(\xi)}{\rho_0} = \frac{\hat{F}_1(1-\hat{F}_{-1}) + (-1)^j\hat{F}_{-1}(1-\hat{F}_1)}{(1-\xi)[1-(1-\sqrt{1-\xi^2})/\xi](1-\hat{F}_1\hat{F}_{-1})}. \tag{11}$$

This result gives access to the full statistics of the position of the TP and puts forward striking characteristics of active transport in dense diffusive single-file systems as detailed below.

(i) The first conclusion we can draw from Eq. (11) is that for arbitrary f (including $f=0$), all odd cumulants have the same generating function $\hat{\kappa}_{\text{odd}}(\xi)$ and all even cumulants have the same generating function $\hat{\kappa}_{\text{even}}(\xi)$. This means that at any moment of time and for any f, all cumulants $\kappa_j^{(n)}$ with arbitrary odd j are equal to each other $\kappa_{2j+1}^{(n)} = \kappa_{\text{odd}}^{(n)}$, and so do all the cumulants with arbitrary even j, $\kappa_{2j}^{(n)} = \kappa_{\text{even}}^{(n)}$.

Parenthetically, we note that, in the classical case of single-file diffusion (i.e., $f=0$), the generating function in Eq. (11) can be inverted explicitly to give $\kappa_{\text{odd}}^{(n)} \equiv 0$ and for arbitrary time moment n

$$\lim_{\rho_0\to 0}\frac{\kappa_{\text{even}}^{(n)}}{\rho_0} = \frac{2}{\sqrt{\pi}}\frac{\Gamma(\lfloor\frac{n-1}{2}\rfloor+\frac{3}{2})}{\Gamma(\lfloor\frac{n-1}{2}\rfloor+1)}, \tag{12}$$

where $\Gamma(\cdot)$ is the Gamma function and $\lfloor x\rfloor$ is the floor function. This expression, which can be shown to be compatible with the well-known Gaussian form in the large time limit, seems to be new.

(ii) Second, turning to the limit $\xi \to 1$ (large-n limit), we find the leading in time asymptotic behavior of the cumulants of arbitrary order:

$$\lim_{\rho_0\to 0}\frac{\kappa_{2j+1}^{(n)}}{\rho_0} = (p_1-p_{-1})\sqrt{\frac{2n}{\pi}} - 2p_1p_{-1}(p_1-p_{-1}) + o(1), \tag{13}$$

$$\lim_{\rho_0\to 0}\frac{\kappa_{2j}^{(n)}}{\rho_0}=\sqrt{\frac{2n}{\pi}}+o(1). \tag{14}$$

Equations (13) and (14) signify that, remarkably, the leading in time behavior of all even cumulants is *independent* of the force f, while the leading in time behavior of all odd cumulants does depend on f. In addition, for the standard choice of the transition probabilities such that $p_1 = 1 - p_{-1}$ and $p_1/p_{-1} = \exp(\beta f)$, where β is the reciprocal temperature, and for the specific case $j = 0$, we check from Eq. (13) that

$$\lim_{\rho_0\to 0}\frac{\bar{X}}{\rho_0}=\tanh(\beta f/2)\sqrt{2n/\pi}, \tag{15}$$

which reproduces, for $j = 0$, the results of Refs. [24,25]. Note that this anomalous scaling $\propto\sqrt{n}$ holds for all cumulants.

(iii) In the specific case of an infinitely strong force f, these large time behaviors can be understood from a simple picture relying on the first-passage time properties of the independent diffusing vacancies obtained above. In this case of a directed motion, the position of the TP is given by the number of times the TP has been visited by any of the vacancies located ahead of it. This quantity is itself easily related to the waiting time distribution between two consecutive visits of a vacancy to the TP. Using the standard tools of the target annihilation problem (see, for instance, Refs. [31,32]), the corresponding survival probability $S(n)$, i.e., the probability that at step n the TP has not been visited by any of the vacancies, can be written as $S(n) = e^{-\rho_0\alpha(n)}$ with

$$\alpha(n)=\sum_{Z=1}^{\infty}F_Z^{(n)}\underset{n\to\infty}{\simeq}\sqrt{\frac{2n}{\pi}}, \tag{16}$$

where we have used the relation (10). Adopting now for the sake of simplicity a continuous time description, the Laplace transform of the renewal process $X(t)$ reads

$$\mathcal{L}\{\Pr[X(t)=m]\}(s)=\hat{S}(s)[1-s\hat{S}(s)]^m, \tag{17}$$

where $\hat{S}(s)$ stands for the Laplace transform of the survival probability [30]. Noticing that Eq. (16) leads to

$$\lim_{\rho_0\to 0}\frac{1-s\hat{S}(s)}{\rho_0}\underset{s\to 0}{\simeq}\frac{1}{\sqrt{2}s^{3/2}}, \tag{18}$$

it is finally found by using Tauberian theorems that

$$\lim_{\rho_0\to 0}\frac{\kappa_j(t)}{\rho_0}\underset{t\to\infty}{\simeq}\sqrt{\frac{2t}{\pi}}, \tag{19}$$

in agreement with the leading contribution of Eqs. (13) and (14) in the specific case $p_1 = 1$. Note that this simple picture where each vacancy that started ahead of the TP interacts at most once with the TP so that all steps of the TP are independent holds only in the regime of infinitely strong force. For a finite force, a given vacancy can visit many times the TP, leading to nontrivial anticorrelation effects quantified by our exact solution.

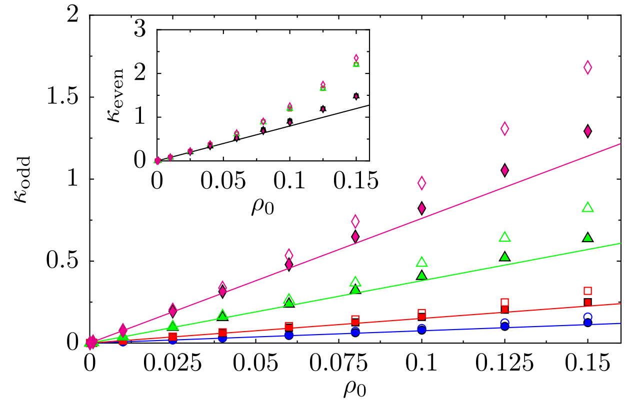

FIG. 2 (color online). Odd cumulants at time $n = 100$ vs ρ_0. The straight lines define our predictions in Eq. (11) for different values of p_1, while the filled and empty symbols are the results of numerical simulations for the first and third cumulants, respectively. Circles are results for $p_1 = 0.55$, squares for $p_1 = 0.6$, triangles for $p_1 = 0.75$, and diamonds for $p_1 = 0.98$. The inset shows analogous results for the second and fourth cumulants.

(iv) We finally provide an explicit expression of the full distribution function $P^{(n)}(X)$ for any n. As a matter of fact, the equality at leading order in ρ_0 of cumulants of the same parity proved in point (i) shows that the distribution associated to these cumulants is of Skellam type [33], so that

$$P^{(n)}(X)\underset{\rho_0\to 0}{\simeq}\exp(-\kappa_{\rm even}^{(n)})\left(\frac{\kappa_{\rm even}^{(n)}+\kappa_{\rm odd}^{(n)}}{\kappa_{\rm even}^{(n)}-\kappa_{\rm odd}^{(n)}}\right)^{X/2}\times I_X\left(\sqrt{(\kappa_{\rm even}^{(n)})^2-(\kappa_{\rm odd}^{(n)})^2}\right), \tag{20}$$

where $I_X(\cdot)$ is the modified Bessel function. Importantly, we find that despite the known asymmetry of the concentration profile of the bath particles [24], the rescaled

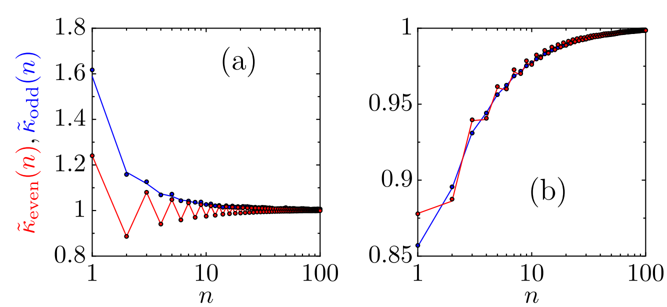

FIG. 3 (color online). Reduced cumulants $\tilde{\kappa}_{\rm even}^{(n)}=\kappa_{\rm even}^{(n)}/\sqrt{2n/\pi}$ and $\tilde{\kappa}_{\rm odd}^{(n)}=\kappa_{\rm odd}^{(n)}/[(p_1-p_{-1})\sqrt{2n/\pi}-2p_1p_{-1}(p_1-p_{-1})]$ vs time n for $\rho_0 = 0.01$ and (a) $p_1 = 0.6$ and (b) $p_1 = 0.98$. Solid lines give the results of the inversion of Eq. (11), while symbols are the results of numerical simulations.

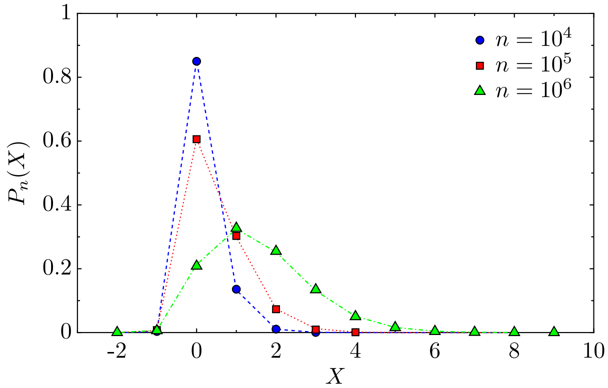

FIG. 4 (color online). The distribution $P^{(n)}(X)$ for $\rho_0 = 0.002$ and $p_1 = 0.98$. The dashed lines are our theoretical predictions in Eq. (20), while the symbols are the results of numerical simulations.

variable $(X - \kappa_{\rm odd}^{(n)})/\sqrt{\kappa_{\rm even}^{(n)}}$ is asymptotically distributed accordingly to a normal law.

Note finally that the regime of validity of our expressions with respect to the density ρ_0 is tested in Fig. 2, where we compare our theoretical predictions for the cumulants, obtained by the inversion of our general Eq. (11), against the results of numerical simulations for different values of the density ρ_0 of the vacancies, for different forces f [defined as $\beta f = \ln(p_1/p_{-1})$] and a fixed time moment $n = 100$. We observe a very good agreement for very small values of ρ_0 and conclude that, in general, the approach developed here provides a very accurate description of the TP dynamics for $\rho_0 \lesssim 0.1$. Further on, in Fig. 3, we plot our theoretical predictions for the time evolution of the cumulants for different values of the force and at a fixed density ρ_0. Again, we observe a perfect agreement between theory and simulations. Note that for small fields, the reduced odd cumulants approach 1 from above while for strong fields from below. Last, we compare in Fig. 4 our prediction in Eq. (20) against the numerical data and again observe a very good agreement between our analytical result and numerical simulations.

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