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Scaling laws in the diffusion limited aggregation of persistent random walkers

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ABSTRACT

We investigate the diffusion limited aggregation of particles executing persistent random walks. The scaling properties of both random walks and large aggregates are presented. The aggregates exhibit a crossover between ballistic and diffusion limited aggregation models. A non-trivial scaling relation $\xi \sim \ell^{1.25}$ between the characteristic size ξ , in which the cluster undergoes a morphological transition, and the persistence length ℓ , between ballistic and diffusive regimes of the random walk, is observed.

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1. Introduction

Aggregation processes are outstanding phenomena that have been subject of theoretical, experimental and simulation investigations in many fields of knowledge [1,2]. The diffusion limited aggregation (DLA) model proposed by Witten and Sander [3] is the most studied theoretical aggregation process. In this model, free particles are released, one at a time, far from a growing cluster and perform successive random jumps while they are not adjacent to the cluster. If a free particle encounters any particle of the cluster, it irreversibly sticks in the corresponding position and becomes part of the cluster. Notwithstanding the rule simplicity, the DLA model produces clusters exhibiting complex scaling properties [4–7]. Another simple and well studied growth process is the so called ballistic aggregation (BA), in which the free particles move ballistically at randomly chosen directions and obey the same sticking rules as the DLA model. The BA model produces asymptotically homogeneous radial clusters [8] whose active (growing) zone is described by the Kardar–Parisi–Zhang (KPZ) universality class [9], as observed in the planar version where the flux of particles is normal to an initially flat substrate [10,11].

Several generalizations of the DLA model have been studied [1] with special attention in those where the free particles perform random walks with a drift [12–15]. In these models, the clusters undergo a crossover from a DLA to a BA scaling regime as the number of particles increases due to the trajectories becoming essentially ballistic at asymptotic large scales. In another group of models, in which the free particles perform long random steps of a fixed length, an inverted transition from BA to DLA is observed [16,17]. In this case, the trajectories are essentially random at asymptotic large scales, independently of the step length. A simple characterization of the morphological transition is done by relating the mass M of an aggregate with its radius r . This quantity has two asymptotic scaling regimes, $M \sim r^2$ and $M \sim r^{1.71}$, related to either the BA and the DLA models, respectively.

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Aggregation models with drift or long steps can be used to investigate systems with a long mean free path as, for example, the aggregation of methane or ammonia molecules in superfluid helium [18]. In particular, an off-lattice DLA model with long steps of length δ was recently applied to the aggregation of molecules in superfluid media [17]. In addition to this example, biased diffusion and super-diffusivity have also been reported in several systems ranging from adatom surface diffusion [19,20] to cell migration [21,22]. The relevance of modelling the aggregation with this kind of random walk is strengthened due to a possible confusion between biased diffusion and super-diffusivity [23].

The representation of a large mean free path by a ballistic movement is a simplifying hypothesis since weak perturbations can promote slight deviations of the ballistic path resulting a biased diffusion at short scales. Motivated by the wide availability of systems for potential applications, we investigate a two-dimensional aggregation process where the particle trajectories consist of random walks, with the direction of a new step limited by an angle δ_θ in relation to the previous step direction. This correlated walk, which was formerly investigated in other contexts [24–26], has a drift at short scales and becomes a random walk at large scales. Also, an aggregation model using this random walk was formerly investigated in a small cluster size limit and a fractal dimension depending on the δ_θ parameter was reported [15]. Actually, this dependence is a finite size effect since this kind of trajectory leads to a BA to DLA crossover, as demonstrated in the present work.

The paper is organized as follows. Model and simulation strategies are presented in Section 2. The trajectories are numerically investigated and an analytical approach for the trajectory crossover is developed in Section 3. In Section 4, the simulations of large clusters are presented and a scaling analysis of the morphological transition is developed. Finally, some conclusions and prospects are drawn in Section 5.

2. Model

We perform two-dimensional off-lattice simulations with an initial cluster consisting of a single particle of diameter a (a seed) stuck to the origin. Free particles are sequentially released, one at a time, at a circle of radius $r_l \gg r_{\max}$ centred at the seed, where r_{\max} is the largest distance from the seed of a particle belonging to the cluster. The free particles follow a persistent random walk [24], in which the position of the n th step is given by

$$\begin{aligned} x_n &= x_{n-1} + a \cos \phi_n \\ y_n &= y_{n-1} + a \sin \phi_n \end{aligned} \quad (1)$$

and the direction of the n th step depends on the preceding one as

$$\phi_n = \phi_{n-1} + \eta_n. \quad (2)$$

The white noise η_n is a random variable uniformly distributed in the interval $(-\delta_\theta/2, \delta_\theta/2)$ and the parameter δ_θ limits the next move direction inside an angular opening of size δ_θ , centred in the direction of the previous step.

The trajectory becomes ballistic for $\delta_\theta \rightarrow 0$ and random for $\delta_\theta \rightarrow 2\pi$ implying in the BA and DLA limit cases, respectively. The direction of the first step is chosen at random. The trajectory is stopped whenever the particle visits a position adjacent to the cluster where it irreversibly sticks. Finally, the particle is discarded whenever it crosses a distance $r_k \gg r_l = r_{\max} + \Delta$. The procedure is repeated while the cluster does not reach N particles. The previously introduced variables r_l and r_k must be as large as possible, but computational limitations restrict their values. For the DLA simulations, Δ can be a few particle diameters [27]. However, for the BA limit, Δ cannot be small due to distortions caused by shadow instabilities [28,29]. The BA clusters are free from shadow effects when $\Delta > 600$ and thus $\Delta = 800$ and $r_k = 10 \times r_l$ were adopted in all simulations.

Since the asymptotic behaviour is typically reached only for large clusters, an efficient algorithm is required. We adopted the strategy where particles execute long steps with the proper angular distribution $P(\phi)$ if they are not close to the cluster [12]. A closed-form expression for the angular distribution of a long jump is not known a priori. So, we numerically computed the probability of the first passage in a circle of radius r occurring at an angle ϕ for a trajectory started at the origin. Fig. 1 shows some angular distributions for different values of δ_θ and r obtained with up to 10^8 independent trajectories. If r and δ_θ are not too large, the distributions are very well fitted by

$$P(\phi) = A \exp(B \cos \phi) + C. \quad (3)$$

Parameters A , B and C (actually only two of them are independent due to the normalization) were determined using non-linear regressions that are also shown in Fig. 1. Cluster growth optimizations were implemented using steps of size $16a$ in the large empty regions nearby the cluster and three step sizes ($16a$, $100a$ and $200a$) were used far from the cluster. More details about long step and off-lattice optimizations are available elsewhere [27].

3. Trajectory scaling properties

In Fig. 2 we show two stages of a trajectory corresponding to short and long times. Roughly, the trajectory consists in a drift around the initial direction until a large angular fluctuation is selected, as illustrated in the left panel of Fig. 2. The smaller angular opening, the rarer the probability of selecting a large fluctuation. However, these rare events unavoidably occur for a sufficiently long time if $\delta_\theta \neq 0$ and the trajectories become erratic at large scales, as shown in the right panel of Fig. 2. In order to investigate this crossover, 10^4 independent trajectories were simulated. In Fig. 3(a) the root-mean-square (RMS) displacement λ is shown for several values of δ_θ . One can clearly resolve a crossover: the RMS displacement scales as $\lambda \sim t$ at early stages and crosses over to a scaling $\lambda \sim t^{1/2}$, as expected in a transition from ballistic to diffusive trajectories.

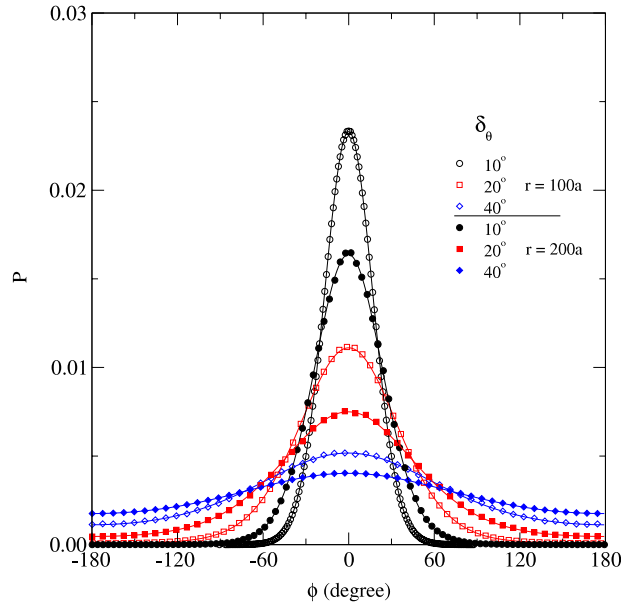


Fig. 1. (Colour on-line) Angular distributions of the first passage at a distance r for a persistent random walker starting at the origin. Without loss of generality, we set $\phi_0 = 0$. Symbols represent stochastic simulations and solid lines represent non-linear regressions using Eq. (3).

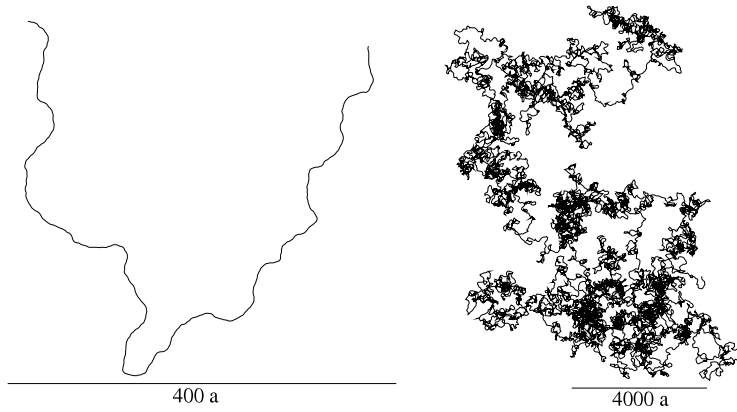


Fig. 2. A typical trajectory for $\delta_\theta = 10^\circ$ after 10^3 (left) and 10^6 (right) steps.

The characteristic crossover time depends on the angular opening as $\tau \sim \delta_\theta^{-2}$. This result can be proved with the following reasoning. Without loss of generality, we assume $\phi_0 = 0$ in Eq. (2) resulting

$$\phi_t = \sum_{k=1}^t \eta_k,$$

where η is a white noise with $\langle \eta \rangle = 0$, $\langle \eta^2 \rangle \propto \delta_\theta^2$, and $\langle \dots \rangle$ means statistical averages. Obviously, the turning angle is a random variable with $\langle \phi_t \rangle = 0$. Applying the central limit theorem [30], in which the sum of t identical and independent random variables with average $\langle \eta \rangle = 0$ and variance $\langle \eta^2 \rangle$ converges to a normal distribution with variance $\sigma_t^2 = t \langle \eta^2 \rangle$, the probability of selecting a large turning angle is exponentially negligible for $\sigma_t \ll 1$ and appreciable for $\sigma_t \gg 1$. Assuming a crossover time given by

$$\sigma_\tau = \sqrt{\tau \langle \eta^2 \rangle} \sim 1,$$

we immediately find out the scaling law $\tau \sim \delta_\theta^{-2}$. Finally, the characteristic RMS displacement at the crossover is proportional to τ since the trajectory is still approximately ballistic at the crossover. These scaling properties λ can be expressed in the following scaling ansatz

$$\lambda(t, \delta_\theta) = t g\left(\frac{t}{\tau}\right), \tag{4}$$

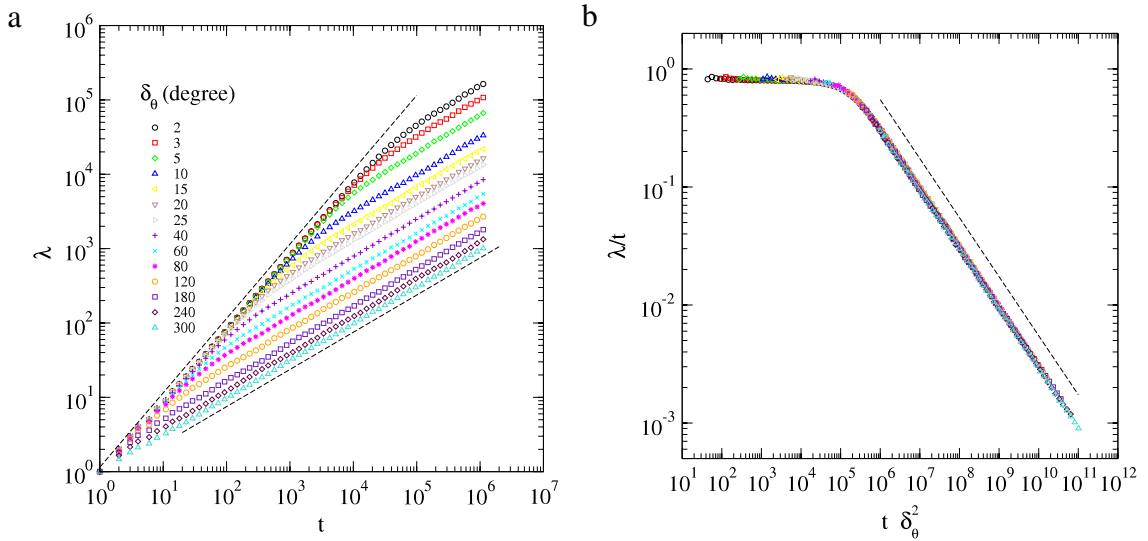


Fig. 3. (Colour on-line) (a) RMS displacements for distinct angular openings. Dashed lines represent scaling laws $\lambda \sim t$ and $\lambda \sim t^{1/2}$. (b) Collapse of curves shown in Fig. 3(a) using the scaling ansatz given by Eq. (4). The dashed line represents the asymptotic behaviour of the scaling function $g(x) \sim x^{-1/2}$.

where the scaling function $g(x)$ has the properties $g(x) \sim \text{const.}$ for $x \ll 1$ and $g(x) \sim x^{-0.5}$ for $x \gg 1$. As one can see in Fig. 3(b), this ansatz collapses the data shown in Fig. 3(a) onto a universal curve if we plot λ/t against $t \times \delta_\theta^2$, demonstrating the ansatz correctness.

Before analysing the aggregates obtained using the persistent random walk, we shortly draw a comparison with the results reported by Tojo and Argyrakis for this persistent random walk [24], in which a scaling $\tau \sim \delta_\theta^{-1.88}$ was numerically obtained in stochastic simulations. This exponent, that is slightly different from ours, results from the crossover times estimated using the intersections between early and long time scaling laws of λ versus t . We instead deduced the scaling behaviours and the theory perfectly matches the numerical simulations as shown in Fig. 3(b).

4. Aggregate simulations and scaling analysis

This section is devoted to the simulations of a DLA model using the persistent random walk defined by Eqs. (1) and (2). Fig. 4 shows different stages of two clusters obtained for angular openings $\delta_\theta = 40^\circ$ and 60° . In both cases, the change of the cluster morphology is evident. At early stages, the aggregates have dense branched morphologies (low density of inner voids) that resemble the BA patterns while sparse branches, the hallmark of DLA clusters, are obtained after a characteristic number of particles. This behaviour qualitatively confirms the morphological crossover predicted with the trajectory properties: the smaller δ_θ , the longer the crossover time. Fig. 5 shows clusters of the same size obtained with distinct δ_θ values. The simulations were stopped when the clusters reached a circle of radius $r = 1000a$. A transition from dense to ramified structures is observed and a fractal dimension dependent on δ_θ can be effectively measured in agreement with Ref. [15]. However, it is important to notice that the dense morphologies are transient and the patterns unavoidably become ramified with the DLA fractal dimension ($D \sim 1.71$) for asymptotically large clusters.

The quantitative characterization of the crossover was done using the mass–radius method [8]. Clusters were grown until they cross a circle of radius $5 \times 10^3 a$. In order to perform statistical averages, 100 independent samples were simulated for each investigated δ_θ value. The mass–radius method consists in determining the number of particles inside a circle of radius r centred in the seed. In general, the mass scales as $M(r) \sim r^D$, where D is the fractal dimension. We have $D \simeq 1.71$ and $D \simeq 2$ for asymptotically large DLA and BA clusters, respectively [8,12].

Fig. 6 shows the mass–radius curves corresponding to different values of δ_θ . The curves have a crossover between the scaling regimes $M \sim r^2$ at small radius and $M \sim r^{1.71}$ at large radius. The mass is described by a scaling form

$$M(r, \delta_\theta) = \delta_\theta^{-2z} f(\delta_\theta^z r), \quad (5)$$

where $f(x) \sim x^2$ for $x \ll 1$ and $f(x) \sim x^{1.71}$ for $x \gg 1$. This scaling ansatz supposes that the crossover radius scales as $\xi \sim \delta_\theta^{-z}$. It also assumes a constraint $M(\xi) \sim \xi^2$ since the clusters are still dense close to the crossover, explaining the exponent $2z$ in Eq. (5). Determining the crossover radius as the intersection between the scaling laws $M_1 \sim r^2$ and $M_2 \sim r^{1.71}$, we obtained a scaling law very close to $\xi \sim \delta_\theta^{-2.5}$. We also evaluated the mass at the crossover and the relation $M(\xi) \sim \xi^2$ was confirmed. The crossover mass and radius against δ_θ are shown in the inset of Fig. 6.

In Fig. 7 the mass–radius curves are collapsed using the scaling ansatz given by Eq. (5) with the exponent $z = 2.5$. As can be seen, an excellent collapse onto the scaling function $f(x)$ is obtained. For the general case with a drift quantified by

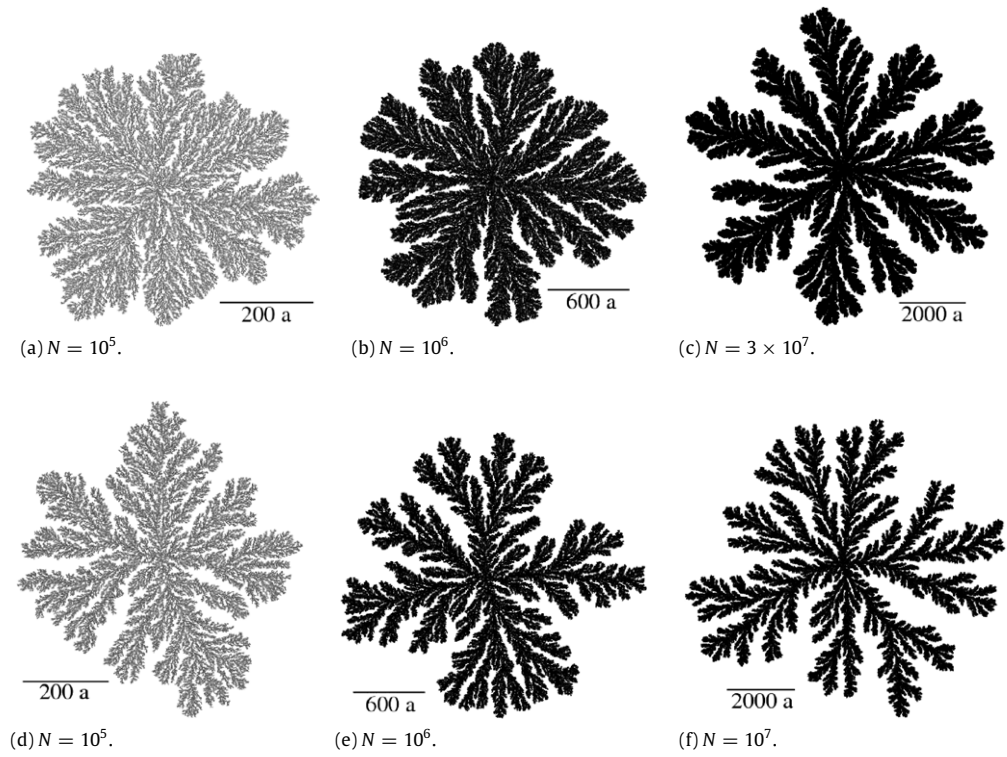


Fig. 4. Different growth stages of clusters generated for $\delta_\theta = 40^\circ$ in (a)–(c) and 60° in (d)–(f). The number of particles are shown below each panel.

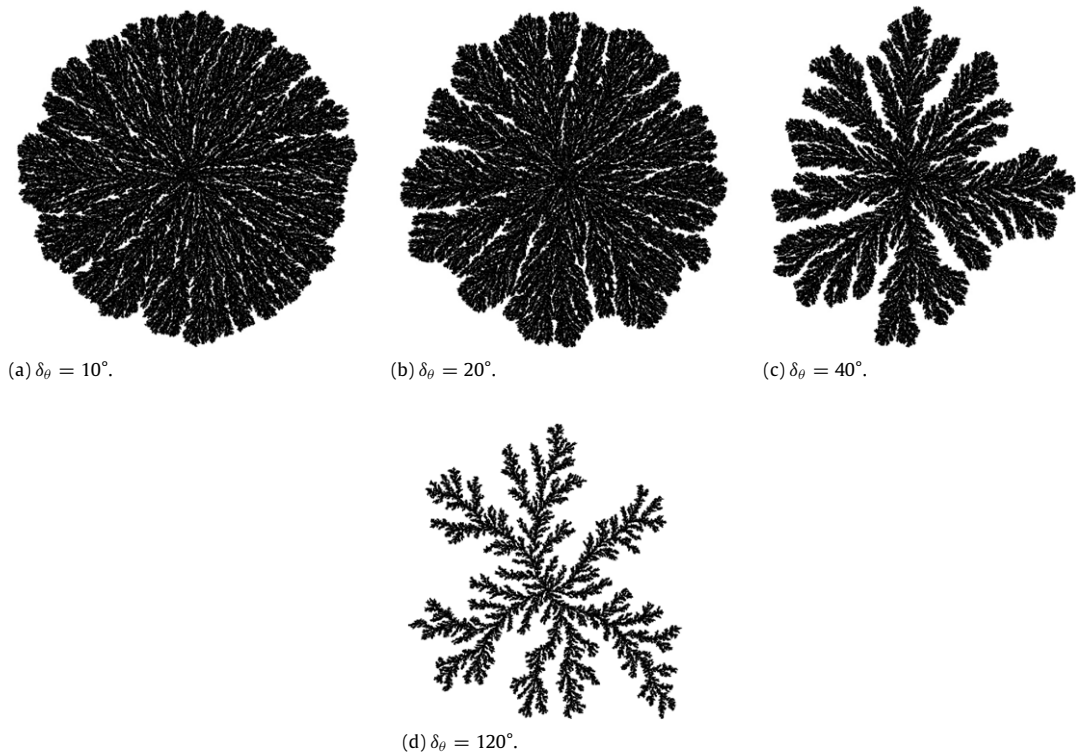


Fig. 5. Clusters of a fixed size $r \approx 1000a$ obtained for distinct δ_θ values. The number of particles varies from 10^4 for the most ramified to 10^6 for the densest cluster.

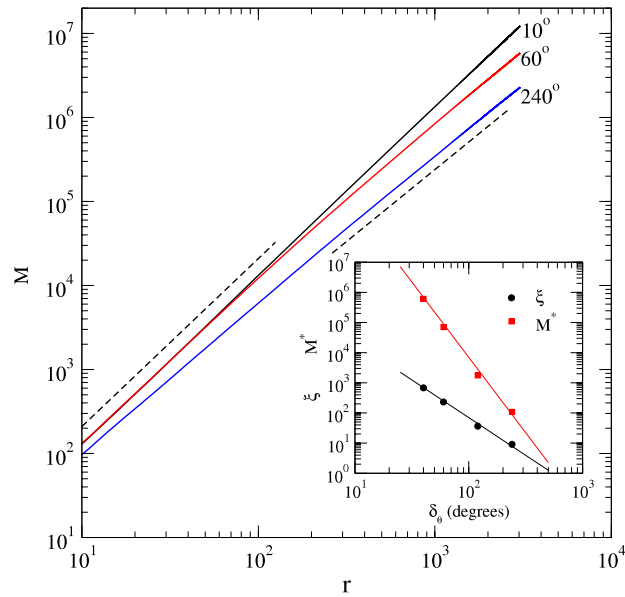


Fig. 6. (Colour on-line) Mass–radius curves for three δ_θ values. Dashed lines are power laws with exponents 2 (smaller r) and 1.71 (larger r) included as guides to the eyes. The inset shows the mass M^* and the characteristic radius ξ at the crossover as functions of δ_θ . Power law regressions with exponents -5 and -2.5 are also shown in the inset.

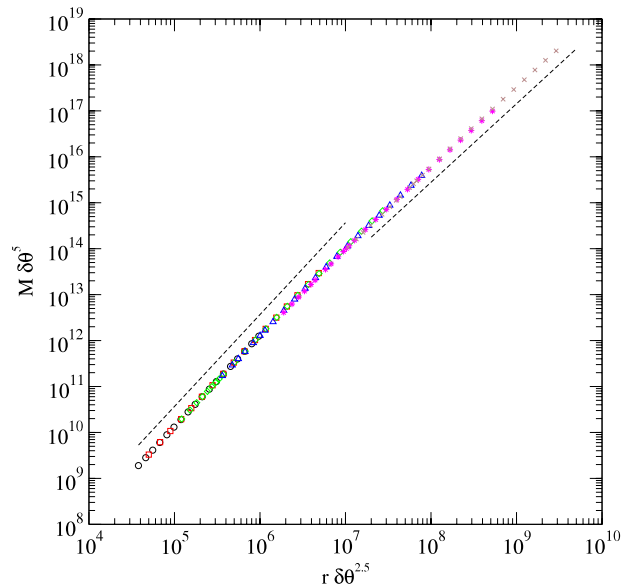


Fig. 7. (Colour on-line) Collapses of the mass–radius curves for $\delta_\theta = 10^\circ, 20^\circ, 40^\circ, 60^\circ, 120^\circ,$ and 240° using the scaling ansatz given by Eq. (5). The solid lines are scaling laws r^2 and $r^{1.71}$ corresponding to the asymptotic behaviour of the scaling function $f(x)$.

a parameter b , the mass against radius can be written as [17]

$$M(r; b) = b^\alpha f\left(\frac{r}{b^\zeta}\right), \tag{6}$$

where the scaling laws $\xi \sim b^\zeta$ and $M(\xi) \sim b^\alpha$ are assumed for the crossover radius and the corresponding mass, respectively. The scaling function $f(x)$ yields the asymptotic regimes of $M(r)$: For a DLA to BA transition, we have $f(x) \sim x^{1.71}$ for $x \ll 1$ and $f(x) \sim x^2$ for $x \gg 1$ while the scaling exponents are exchanged to $f(x) \sim x^2$ for $x \ll 1$ and $f(x) \sim x^{1.71}$ for $x \gg 1$ for a BA to DLA transition. We can easily relate the scaling hypothesis (5) with the scaling ansatz given by Eq. (6) for the aggregation of particles performing long steps of size $\ell = b$. It was shown in Section 3 that the characteristic length in a persistent random walk scales as $\ell \sim \delta_\theta^{-2}$. Replacing $\delta_\theta \sim 1/\sqrt{\ell}$ in Eq. (5) and comparing with Eq. (6), we find the exponents $\alpha = z = 2.5$ and $\zeta = z/2 = 1.25$. These exponents are consistent with those obtained for long steps trajectories in Ref. [17] indicating a universality class.

5. Conclusions

In this work we have investigated the scaling properties of a diffusion limited aggregation (DLA) model where the particles follow locally persistent trajectories. The bias is introduced by limiting the direction of a new step into an angular opening δ_θ in relation to the preceding step direction. We show analytically and numerically that this trajectory is effectively ballistic below a length scale $\ell \sim \delta_\theta^{-2}$ and becomes random at large scales. This result improves the numerical estimate of the scaling law reported formerly by Tojo and Argyrakis [24].

Large scale simulations show that the aggregates undergo a morphological transition from a ballistic aggregation (BA) regime at small sizes to a DLA-like ramified morphology at asymptotically large scales. It is important to mention that a previous analysis of this aggregation model [15], which was done in a small cluster size limit, states a non-universal fractal dimension. However, we have shown that the asymptotic behaviour of the clusters is described by the DLA fractal dimension. Finally, the crossover between the BA and DLA scaling regimes is related with the angular opening by $\xi \sim \delta_\theta^{-2.5}$ indicating a non-trivial relation between the characteristic size of the cluster in the crossover and the persistence length of the random walk.

The persistent random walk has interesting features that can be associated to a number of physical systems [18,22,26,31,32]. A small angular opening may represent a diffusion in a medium where the scattering is weak except by rare but no negligible fluctuations. An important example is the wandering/aggregation of molecules in superfluid helium [18] where the particles may have a mean free path ranging from a molecule diameter to micrometres depending on the proximity to the critical temperature. This kind of diffusion is also observed in cell migration [21,22], a chemotactic process in which the cells diffuse following a gradient-mediated chemical signalling. A relevant property of the trajectory is a characteristic mean free path or persistence length that emerges naturally as we have demonstrated with the central limit theorem [30]. In the cited examples, the aggregation and the consequent formation of clusters were subjects of recent experimental interest [18,33,34]. Consequently, the present theoretical analysis can be extended to future applied investigations.

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