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# On the non-equilibrium phase transition in evaporation–deposition models

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**Abstract.** We study a system of diffusing–aggregating particles with deposition and evaporation of monomers. By combining theoretical and numerical methods, we establish a clearer understanding of the non-equilibrium phase transition known to occur in such systems. The transition is between a growing phase in which the total mass increases for all time and a non-growing phase in which the total mass is bounded. In addition to deriving rigorous bounds on the position of the transition point, we show that the growing phase is in the same universality class as diffusion–aggregation models with deposition but no evaporation. In this regime, the flux of mass in mass space becomes asymptotically constant (as a function of mass) at large times. The magnitude of this flux depends on the evaporation rate but the fact that it is asymptotically constant does not. The associated constant flux relation exactly determines the scaling of the two-point mass correlation function with mass in all dimensions while higher order mass correlation functions exhibit nonlinear multi-scaling in dimension less than two. If the deposition rate is below some critical value, a different stationary state is reached at large times characterized by a global balance between evaporation and deposition with a scale-by-scale balance between the mass fluxes due to aggregation and evaporation. Both the mass distribution and the flux decay exponentially in this regime. Finally, we develop a scaling theory of the model near the critical point, which yields non-trivial scaling laws for the critical two-point mass correlation function with mass. These results are well supported by numerical measurements.

**Keywords:** exact results, irreversible aggregation phenomena (experiment), stochastic particle dynamics (theory), turbulence

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## 1. Introduction

Simple lattice-based particle systems undergoing diffusion and irreversible aggregation provide a useful approach to modelling various phenomena in physics such as polymer growth kinetics, atmospheric aerosol formation and thin film growth on substrates. For a review, see [1] and the references therein. Through various mappings and analogies, diffusion–aggregation models can be connected to a quite diverse range of topics in statistical physics such as the geometry of river networks [2], self-organized criticality [3] and force fluctuations in granular bead packs [4]. It has also been long recognized [5] that such models, in the presence of an external source of monomers, also provide excellent examples of systems which reach a non-equilibrium stationary state with a flux of a conserved quantity (in this case, mass) and exhibit scaling behaviour far from equilibrium. The simplest such model, and the basis for much of what follows, is Scheidegger’s river network model [6] or the Takayasu model [7]–[9] in which particles diffuse with mass independent diffusion rate, and coagulate on contact conserving mass, with input of particles from outside. Much is known about the statistics of this model (see [10, 11] for some more recent developments). In particular, it always reaches a stationary state and has critical dimension,  $d_c = 2$ . In dimension  $d > 2$  the Smoluchowski mean field theory is applicable and the stationary cluster mass distribution scales as  $m^{-3/2}$ , whereas for  $d < 2$  diffusive fluctuations become essential and the scaling of the cluster mass distribution is modified to  $m^{-(2d+2)/(d+2)}$ . In all cases the stationary state is characterized by a constant flux of mass from small to large masses. As a consequence, the scaling exponent of the two-point mass correlation function responsible for the mass flux (to be defined later) can be determined exactly in all dimensions, a result we refer to as a constant flux relation (CFR) [12].

In this paper we are interested in studying what happens when monomers are allowed to evaporate from clusters. By allowing evaporation, mass is no longer strictly conserved. Several questions then arise which we attempt to answer in this paper. Does evaporation change the character of the stationary state? In particular, what happens to the CFR? The effect of evaporation was considered in [13] in a closed system without injection. In that study, evaporating monomers remained in the system acting as an effective source so that the total mass was exactly conserved. The effect of evaporation in an open system (in the sense that evaporating monomers leave the system) with an external source of monomers was studied in [14]. As a result of these studies, it was understood that the system exhibits a non-equilibrium phase transition as the rate of evaporation of monomers is varied. For weak evaporation, the constant flux stationary state persists, albeit with a modified mass flux. In [15] we gathered numerical evidence that the CFR scaling exponent was insensitive to evaporation in this regime. In what follows, we make this observation precise. This regime does not, however, persist for all values of the evaporation rate. Above a certain critical value of the evaporation rate, it becomes impossible to generate large clusters and the mass flux decays exponentially with cluster mass. In this regime, we show that the CFR is replaced by a scale-by-scale balance between the aggregation and evaporation fluxes. In this paper we refer to these two regimes as the *growing phase* and the *exponential phase* respectively.

The connection between the flux of a conserved quantity in a non-equilibrium steady state and universal aspects of the underlying statistics has been an important theme in the theory of fluid turbulence (see [16]–[18] for a review). Kolmogorov’s famous 1941 paper identified the flux of energy through scales of a turbulent flow as the single most important feature of a turbulent steady state. In particular, for stationary, isotropic turbulence, the constant flux of energy exactly determines the scaling of the third order structure function of velocity field which is responsible for carrying the energy flux. This statement is known as the Kolmogorov  $\frac{4}{5}$ -law. It states that for length scales,  $r$ , in the inertial range, the third order structure function is:

$$\langle [v_l(\mathbf{r}) - v_l(\mathbf{0})]^3 \rangle = -\frac{4}{5} \epsilon r, \quad (1)$$

where  $\epsilon$  is the energy flux, and  $v_l$  is the longitudinal component of the velocity. The above law does not depend on the dimensionality of space or details of energy injection and dissipation. This is an important benchmark in the theory of turbulence for which no controlled approximation schemes are known for the computation of general correlation functions. Analogous results have been obtained for other hydrodynamic turbulent systems, such as magneto-hydrodynamics [19], Burgers turbulence [20] and turbulent advection (see [21] and references within). If, based on the  $\frac{4}{5}$ -law, one postulates, as Kolmogorov did in his 1941 theory, that *all* velocity correlation functions depend only on the energy flux as a parameter, then one is led to predict that the  $n$ th order structure function should scale with exponent  $n/3$ . In fact, experiments indicate that turbulence exhibits multi-scaling: the scaling exponent of the  $n$ th order structure function varies nonlinearly with  $n$ . As was realized later by Landau, Kolmogorov’s 1941 theory is an example of ‘mean field’ theory which is valid only if energy flux fluctuations around the mean value are small.

The relevance of this discussion of universal features of turbulence for the present paper, ostensibly on a rather different topic, is outlined in a recent series of

papers [11, 22, 12] in which we explore the implications of the analogy between energy cascades in turbulence and mass cascades in the Takayasu model. We make extensive use, in the present work, of the universal scaling properties of the flux-carrying correlation function as expressed in the CFR [12]—the direct analogue for the Takayasu model of the  $\frac{4}{5}$ -law. For a more detailed discussion of the CFR in the context of cluster aggregation see [23].

The rest of the paper is organized as follows. We first (section 2) define the model and briefly summarize known results on the phase transition and the behaviour of correlation functions in different phases. We then (section 3) use the first equation in the BBGKY hierarchy (Hopf equation) relating the one- and two-point functions of the model to prove the existence of the growing phase of the model and to establish a rigorous upper bound on the position of the transition point. The stationary Hopf equation (at  $t = \infty$ ) is then re-expressed (section 4) as the balance condition between processes of aggregation, deposition and evaporation. The implications of this balance condition are then analysed in the growing phase (section 5) and in the exponential phase (section 6). The existence of an asymptotically constant flux of mass in the growing phase is established and used, in conjunction with the CFR, to calculate the universal scaling of the two-point function in all spatial dimensions. In the exponential phase, the flux decays exponentially and, using a combination of mean field approximation and numerical methods, we show that the CFR is replaced by a scale-by-scale balance between mass fluxes due to evaporation and aggregation. Finally, (section 7) the scaling theory of the model near the critical point is developed, again making use of the CFR to obtain a relation between scaling exponents, and applied to the calculation of correlation functions at the critical point.

## 2. The model

Consider a  $d$ -dimensional hyper-cubic lattice. A lattice site may be occupied by at most one particle. Let the mass of the particle at site  $i$  be denoted by  $m_i$ , where  $m_i = 0, 1, 2, \dots$ . By convention,  $m_i = 0$  corresponds to an empty site. Starting from an initial state where all the lattice sites are empty, the system evolves in time through the following Markovian stochastic processes.

- (i) *Diffusion–coagulation*: with rate  $D$ , a particle hops to one of its randomly chosen nearest neighbour sites. If the target site is already occupied, then the two particles coagulate to form one particle whose mass is the sum of the two particles. If  $i$  is the site and  $j$  the target site, then  $m_i \rightarrow 0; m_j \rightarrow m_j + m_i$ .
- (ii) *Deposition*: with rate  $q$ , a particle of mass 1 is adsorbed at a site  $i$ , increasing the mass at the site by one, i.e.,  $m_i \rightarrow m_i + 1$ .
- (iii) *Evaporation*: with rate  $p$  the mass at a site is decreased by one, provided there is a non-zero mass at that site, i.e.,  $m_i \rightarrow m_i - 1 + \delta_{m_i,0}$ .

By an appropriate renormalization of the timescale, the hopping rate  $D$  is set equal to 1. The model is then characterized by two independent parameters:  $p$ , the evaporation rate, and  $q$ , the deposition rate. We refer to the model as the evaporation–deposition model or EDM.

Let  $P(m, t)$  be the probability that a site is occupied by a particle of mass  $m$  at time  $t$ . More generally, let  $P(m_{i_1}, m_{i_2}, \dots, m_{i_n}; t)$  be the  $n$ -point joint probability distribution

that lattice sites  $i_1, i_2, \dots, i_n$  are occupied by particles with masses  $m_{i_1}, m_{i_2}, \dots, m_{i_n}$  at time  $t$ . In the limit of large time, the EDM approaches a steady state. All probability distribution functions accordingly become independent of time. We will refer to the probability distribution functions in the steady state by dropping the time argument, e.g.,  $P(m) = \lim_{t \rightarrow \infty} P(m, t)$ .

A striking feature of the EDM is the existence of a non-equilibrium phase transition at  $t = \infty$  in all dimensions. This was established in [14] using a combination of mean field analysis and numerical simulations. For fixed evaporation rate  $p$  and sufficiently small deposition rate  $q$  the steady state has finite total mass  $\langle m \rangle = \sum_{m=1}^{\infty} mP(m)$ . If however,  $q$  is increased beyond a certain critical value,  $q_c(p)$ , the total mass in the steady state becomes infinite. It has been also observed that for  $q < q_c(p)$  the large mass asymptotics of  $P(m)$  are exponential, whereas for  $q = q_c(p)$  and  $q > q_c(p)$  the asymptotics of the mass distributions are algebraic but with different exponents:

$$P(m) \sim \begin{cases} e^{-m/m^*} & \text{when } q < q_c, \\ m^{-\tau_c} & \text{when } q = q_c, \\ m^{-\tau} & \text{when } q > q_c, \end{cases} \quad (2)$$

where  $m^*$  is a  $q$  dependent cut-off, and  $\tau \neq \tau_c$ , see [14, 24, 25] for more details. The three phases of the EDM at  $t = \infty$  will be called the exponential phase ( $q < q_c$ ), the critical phase ( $q = q_c$ ) and the growing phase ( $q > q_c$ ).

### 3. The phase transition

The EDM is a Markov model with configuration space  $\mathbf{Z}_+^d$ . Accordingly, the evolution of the probability measure on the configuration space is governed by the master equation. Marginalizing the master equation over all but one lattice site, it is straightforward to derive the system of equations describing the time evolution for  $P(m, t)$ :

$$\begin{aligned} \frac{dP(m, t)}{dt} = & -D \sum_{m'=1}^{\infty} P(m, m', t) - DP(m, t) + D \sum_{m'=0}^{m-1} P(m', m - m', t) \\ & + q[P(m - 1, t) - P(m, t)] + p[P(m + 1, t) - P(m, t)], \quad m > 0, \end{aligned} \quad (3)$$

$$\frac{dP(0, t)}{dt} = -D \sum_{m'=1}^{\infty} P(0, m', t) + D \sum_{m'=1}^{\infty} P(m', t) - qP(0, t) + pP(1, t), \quad (4)$$

where  $P(m_1, m_2, t)$  is the joint probability distribution function of two neighbouring sites having masses  $m_1$  and  $m_2$ . The hopping rate  $D$  will be set equal to one. The simplest way to verify (3) and (4) is to enumerate the number of ways a site can be occupied by a particle of mass  $m$  and the number of ways a site already occupied by a particle of mass  $m$  can change to another mass in a small time  $dt$ . These equations are not closed: they are the evolution equations for the one-point distribution of mass, but their right hand sides contain the two-point joint distribution function. In fact, (3) and (4) are the first members of the infinite BBGKY (Hopf) hierarchy, which in general is impossible to solve without resorting to closure approximations such as mean field theory or perturbative renormalization group. We also point out that the evolution equations for joint probability distribution functions will involve the dimension  $d$ .

There is, nevertheless, a great deal of information about phases of EDM which can be extracted from the first members of the hierarchy without resorting to any closure assumptions. For example, in the growing phase of the EDM, there is a hidden conserved quantity—the flux of mass due to aggregation—which will allow us to extract the exact large mass asymptotics of the two-point mass distribution from (3) and (4) in all space dimensions.

We start by demonstrating the existence of the growing phase of EDM in all dimensions without resorting to any closure approximations such as mean field theory. Multiplying both sides of (3) with  $m$  and integrating over all masses, it is easy to find that  $J(t)$ , the flux of mass into the system, is given by

$$J(t) \equiv \frac{d\langle m(t) \rangle}{dt} = q - ps(t), \tag{5}$$

where  $s(t) = \sum_{m=1}^{\infty} P(m, t)$  is the probability that a site is not empty and  $\langle m(t) \rangle$  is the average mass at time  $t$ . As  $s(t) \leq 1$ ,

$$J(t) \geq q - p. \tag{6}$$

Therefore, if the deposition rate  $q$  is larger than the evaporation rate  $p$ , then the flux of mass into the system is positive at all times, implying that the system is in the growing phase with  $\langle m(\infty) \rangle = \infty$ . Equation (6) provides a simple upper bound for the critical value:  $q_c(p) < p$ .

The above upper bound can be improved quite easily. Equation (4) for  $m = 0$  may be re-written as

$$\frac{dP(0, t)}{dt} = 1 - (2 + q)P(0, t) + pP(1, t) + P(0, 0, t). \tag{7}$$

Dropping the non-negative terms  $pP(1, t)$  and  $P(0, 0, t)$ , we obtain the inequality

$$\frac{dP(0, t)}{dt} \geq 1 - (2 + q)P(0, t), \tag{8}$$

for all times  $t > 0$ , with the initial condition  $P(0, t = 0) = 1$ . This reduces to the following bound on  $P(0, t)$ :  $P(0, t) \geq F(t)$ , where  $F(t)$  is the solution of the linear differential equation

$$\frac{dF(t)}{dt} = 1 - (q + 2)F(t), \quad F(0) = 1. \tag{9}$$

Solving the differential equation, we conclude that

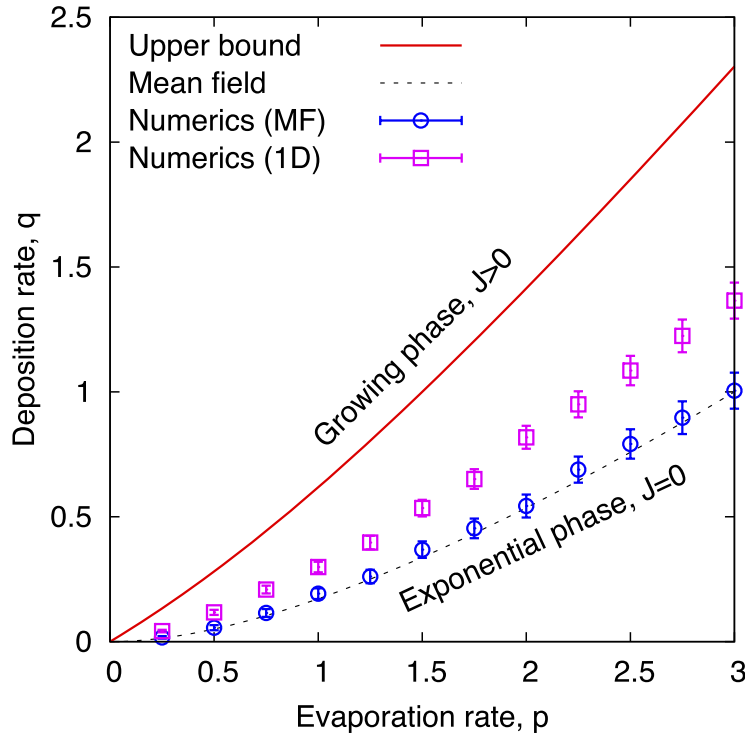
$$P(0, t) \geq \frac{1}{q + 2} + \frac{q + 1}{q + 2} e^{-(q+2)t}. \tag{10}$$

The derived inequality immediately leads to an upper bound on particle density  $s = 1 - P(0, t = \infty)$ :

$$s \leq \frac{q + 1}{q + 2}, \tag{11}$$

or equivalently a lower bound on the mass flux,

$$J = q - ps \geq \frac{q^2 + (2 - p)q - p}{q + 2}. \tag{12}$$



**Figure 1.** Phases of the evaporation–deposition model. Squares: numerical measurement of  $q_c(p)$  in one dimension; dashed curve: mean field result (14); solid curve: upper bound (13); circles: Monte Carlo simulation of the one-dimensional EDM with long range hopping.

The minimum value of  $q$  for which the lower bound for  $J$  is positive is an upper bound for the transition point. Thus, we obtain

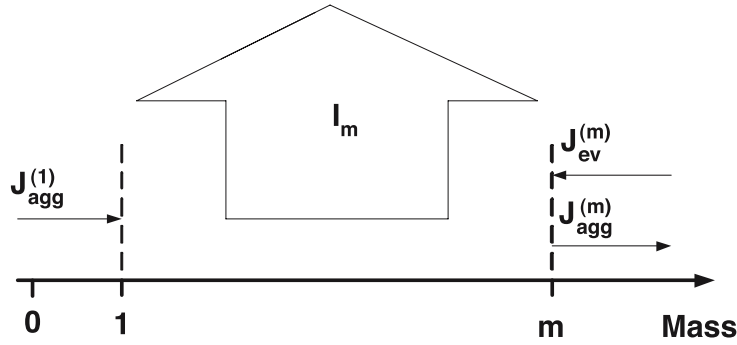
$$q_c^{UB} = \frac{1}{2} \left( p - 2 + \sqrt{p^2 + 4} \right), \quad (13)$$

which establishes an upper bound on  $q_c(p)$  that is tighter than the simple  $q_c(p) < p$  bound.

Proving the existence of the exponential phase or equivalently deriving a lower bound for  $q_c(p)$  is difficult. However, numerical simulations and a non-trivial mean field analysis carried out in [14] show that as we decrease evaporation rate  $q$  while keeping  $p$  constant, the system undergoes a non-equilibrium phase transition into an exponential phase in which the total mass present in the system remains finite at all times. The transition occurs at some critical value of the deposition rate  $q = q_c(p) > 0$ . Within the mean field approximation [14]

$$q_c^{MF}(p) = p + 2 - 2\sqrt{p + 1}. \quad (14)$$

Numerics (see figure 1) suggest that the curve  $q = q_c^{MF}(p)$  is a lower bound on the true phase transition boundary. We cannot prove this conjecture and thus establish the existence of the exponential phase by using just the first pair of equations from the BBGKY (Hopf) hierarchy. The relationship between the rigorous upper bound (13), the mean field estimate of the phase boundary (14) and the numerical estimate of  $q_c(p)$  in one dimension is illustrated in figure 1. We now discuss the large time statistics of the model in each of its phases.



**Figure 2.** Balancing fluxes in the steady state of the model.

#### 4. Mass balance in the steady state

In the steady state the amount of mass contained in every finite interval  $[1, m]$  is constant:

$$\frac{d}{dt} \sum_{\mu=1}^m \mu P(\mu) = 0. \quad (15)$$

Using (3) this can be re-written as the following balance condition:

$$I_m + J_{\text{agg}}^{(m)} = J_{\text{agg}}^{(1)} + J_{\text{ev}}^{(m)}, \quad (16)$$

where

$$I_m = p \sum_{\mu=1}^m P(\mu) - q \sum_{\mu=1}^{m-1} P(\mu) \quad (17)$$

is the bulk term equal to the rate at which mass escapes from the system due to evaporation and deposition,

$$J_{\text{agg}}^{(1)} = qP(0) \quad (18)$$

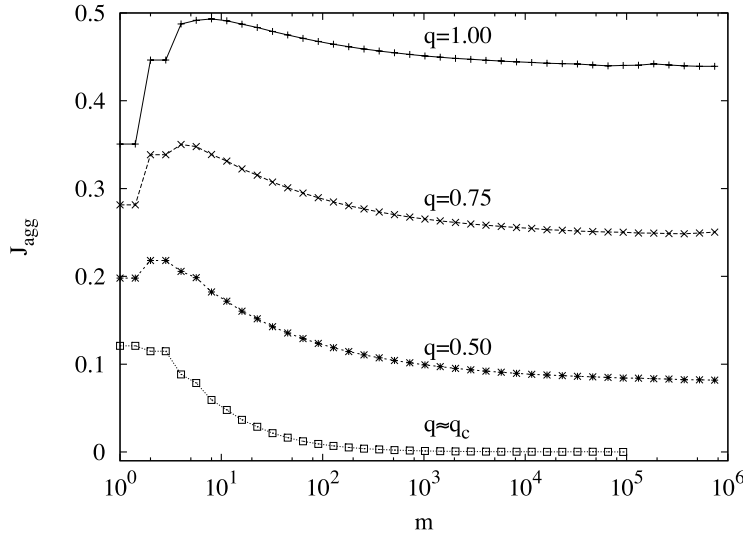
is the flux of mass into the interval  $[1, m]$  due to deposition of particles of unit mass onto empty sites,

$$J_{\text{ev}}^{(m)} = pmP(m+1) \quad (19)$$

is the flux of mass into the interval  $[1, m]$  due to evaporation of particles of mass  $m+1$ , and finally

$$J_{\text{agg}}^{(m)} = 2 \sum_{\mu=1}^m \mu P(\mu) - \sum_{\mu=0}^m \sum_{\mu'=0}^{\mu} \mu P(\mu', \mu - \mu') + qmP(m) \quad (20)$$

is the flux of mass out of the interval  $[1, m]$  due to diffusion–aggregation. Figure 2 illustrates the balance equation graphically. The balance (16) is realized differently in different phases of the model, which is elaborated upon below.



**Figure 3.** Numerical measurement of aggregation flux in the growing and critical phases of the EDM in one dimension. The simulations are for  $p = 1.0$ , when  $q_c \approx 0.3072$ . For  $q > q_c(p)$ ,  $J_{\text{agg}}(m) \xrightarrow{m \rightarrow \infty} J > 0$ . At the critical point, the large mass limit of  $J_{\text{agg}}$  is zero.

## 5. Growing phase

Consider the large mass limit of (16). Being a probability distribution,  $P(m)$  sums to one, which means that  $P(m)$  must go to zero faster than  $1/m$  as mass  $m$  tends to infinity. Therefore,  $J_{\text{ev}}^{(m)}$  vanishes for large masses,

$$J_{\text{ev}}^{(m)} = pmP(m+1) \xrightarrow{m \rightarrow \infty} 0. \quad (21)$$

The bulk evaporation term  $I_m$  approaches a limit:

$$I_m \xrightarrow{m \rightarrow \infty} (p - q)s, \quad (22)$$

where  $s = 1 - P(0)$ . Therefore the balance equation acquires the form

$$J_{\text{agg}}^{(m)} = J_{\text{agg}}^{(1)} - I_{\infty} + O(m^{-\alpha}) = J + O(m^{-\alpha}), \quad \forall m \gg 1, \quad (23)$$

where  $\alpha > 0$  and  $J$  is the total flux of mass at  $m = \infty$ ,

$$J = q - ps, \quad (24)$$

as in (5). In the growing phase  $J > 0$  and we find that the flux of mass due to diffusion–aggregation is asymptotically constant in the limit of large mass despite evaporation and deposition being present at all scales. In figure 3, the variation of  $J_{\text{agg}}^{(m)}$  with  $m$  is shown for different values of  $q \geq q_c$ . For large  $m$ , the current goes to a non-zero constant in the growing phase.

We conclude that the large mass limit of  $J_{\text{agg}}^{(m)}$  in EDM is identical to that of the model with no evaporation, but with a renormalized deposition rate  $J = q - ps$ . The fact that there is a constant flux of mass in the model allows us to apply the constant flux relation derived in [12] to determine the exact scaling of the two-point function in the

growing phase of the model. In particular, the equation  $J_{\text{agg}}^{(m)} = \text{constant}$  for large  $m$  may be solved using the Zakharov transform [12] to obtain

$$P(m_1, m_2) = \frac{1}{(m_1 m_2)^{3/2}} f\left(\frac{m_1}{m_2}\right), \quad m_1, m_2 \gg 1, \quad (25)$$

where  $f(x) = f(1/x)$  is an unknown scaling function. This result holds in all dimensions and is derived without any closure assumptions. We conclude that the two-point function in the growing phase of the EDM scales exactly as in the aggregation model with deposition and no evaporation.

It is worth noting an alternative representation of the above result in Fourier space. Let

$$\psi(z_1, z_2) = \sum_{m_1, m_2=0}^{\infty} P(m_1, m_2) (z_1^{m_1} - 1) (z_2^{m_2} - 1). \quad (26)$$

Then the equivalent form of (25) in terms of  $\psi(z_1, z_2)$  is

$$\psi(z, z) = -J(z - 1) + O[(z - 1)^2], \quad (27)$$

which is strongly reminiscent of the Kolmogorov  $\frac{4}{5}$ -law in fluid turbulence. The easiest way to derive (27) is to differentiate the generating functional  $\psi(z) = \sum_{m=0}^{\infty} P(m) z^m$  with respect to time and resolve the resulting time derivative using the equations of motion (3).

Equation (25) is a direct consequence of the asymptotic constant flux condition (23) which holds in the growing phase despite the presence of evaporation. It is natural to conjecture that the growing phase of the EDM is equivalent to the aggregation model with a suitably renormalized deposition rate. A consequence of such a conjecture would be the expression for all scaling exponents for multi-point probability distribution functions of the model in one dimension. Namely, based on the results of [11], we expect that

$$P(\Lambda m_1, \Lambda m_2, \dots, \Lambda m_n) \sim \Lambda^{-\gamma_n} P(m_1, m_2, \dots, m_n), \quad (28)$$

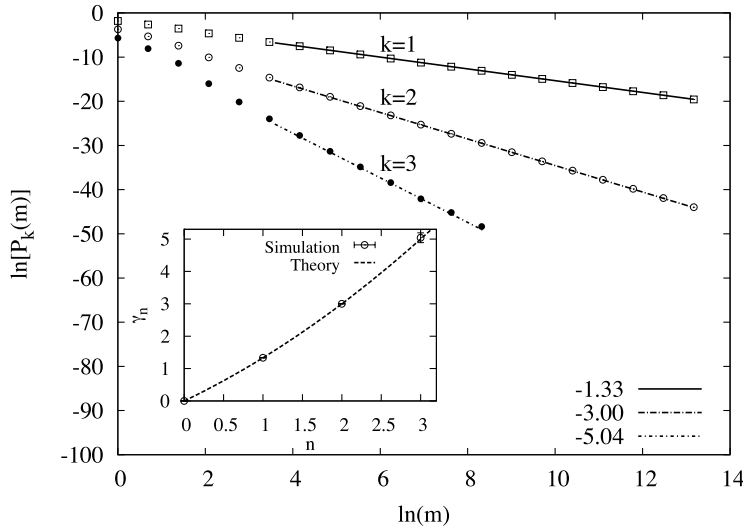
where in one dimension

$$\gamma_n = \frac{4n}{3} + \frac{n(n-1)}{6}, \quad n = 1, 2, 3, \dots, d = 1. \quad (29)$$

We check this conjecture numerically. Let

$$P_k(m) = P(\underbrace{m, m, \dots, m}_k), \quad (30)$$

where the masses  $m$  are on neighbouring sites. We measure  $P_k(m)$  in the growing phase in one dimension. The results are shown in figure 4. The data are in good agreement with (29). A rigorous explanation of the observed result that the growing phase of the aggregation model with evaporation rate  $p > 0$  and deposition rate  $q > 0$  is equivalent to the aggregation model with no evaporation and deposition rate  $q' = J = q - ps$  remains an open problem.



**Figure 4.** Numerical verification of (28) and (29) for one-, two- and three-point correlation functions in one dimension. The theoretical values of the scaling exponents are given by (29):  $\gamma_1 = \frac{4}{3}$ ,  $\gamma_2 = 3$ ,  $\gamma_3 = 5$ .

### 6. Exponential phase

We now analyse the implications of the balance equation (16) in the exponential phase. In this case the total flux of mass at infinite mass is zero. As a consequence, we claim that the large mass behaviour of the model in the exponential balance is described by a steady state characterized by a scale-by-scale balance between aggregation and evaporation fluxes. As far as we are aware, the non-perturbative analysis of the exponential has never been attempted. Therefore, we will base our arguments mostly on mean field results and numerical simulations in one dimension.

The mass fluxes  $J_{ev}^{(m)}$ ,  $I_m$  and  $J_{agg}^{(1)}$  defined in (17)–(19) can be written for large masses as

$$J_{ev}^{(m)} \approx pmP(m), \tag{31}$$

and

$$J_{agg}^{(1)} - I_m \approx (q - ps) - (q - p) \int_m^\infty d\mu P(\mu) = (p - q) \int_m^\infty d\mu P(\mu), \tag{32}$$

where the global mass flux  $q - ps$  has been set to zero for the exponential phase. Clearly, if  $P(m)$  goes to zero at large masses faster than a power law, for example as a stretched exponential or as an exponential, then  $J_{ev}^{(m)} \gg J_{agg}^{(1)} - I_m$ .

It has been shown in [14] that, within the mean field theory, for large masses,  $P(m)$  has the form

$$P(m) \sim \frac{A}{m^{3/2}} e^{-m/m^*}, \quad m \gg 1, \tag{33}$$

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where  $m^*$  is the effective mass determined by the singularities of the mean field moment generating function. It follows that

$$J_{ev}^{(m)} \sim pm^{-1/2}e^{-m/m^*}, \quad (34)$$

and

$$J_{agg}^{(1)} - I_m \sim A(p - q) \frac{m^*}{m^{3/2}} e^{-m/m^*}. \quad (35)$$

Thus,  $J_{agg}^{(1)} - I_m$  decays with mass faster than the flux due to evaporation. The balance equation (16) then reduces to

$$J_{agg}^{(m)} \approx J_{ev}^{(m)}, \quad m \gg 1. \quad (36)$$

Therefore, a scale-by-scale balance is established in the exponential phase for each value of  $m$ . The flux of mass out of the interval  $[1, m]$  due to diffusion–aggregation is equal to the flux of mass into the interval  $[1, m]$  due to evaporation of particles of mass  $m + 1$ . Simultaneously with this scale-by-scale balance, the global balance holds: the rate of the total mass increase due to deposition is exactly equal to the total rate of mass loss due to evaporation. Using the explicit expression (19) for  $J_{ev}^{(m)}$  it is possible to re-write (36) as a relation between mass distribution and the aggregation flux:

$$P(m + 1) \approx \frac{1}{pm} J_{agg}^{(m)}, \quad (37)$$

which replaces the CFR of the growing phase.

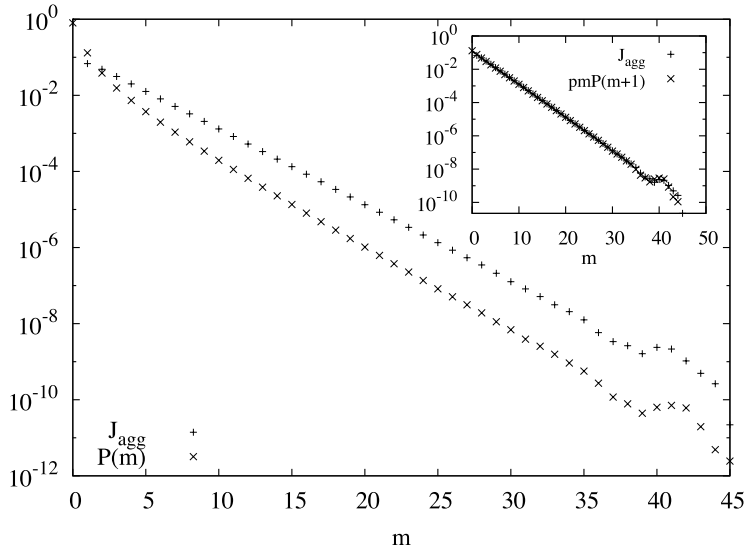
Our conclusion that in the absence of global flux the steady state is described by a scale-by-scale balance between evaporation and aggregation seems natural and should hold irrespective of the validity of the mean field assumption. The exponential nature of  $P(m)$  should be true in one dimension too, though corrections to it would be different from the mean field result. In figure 5, we show the results of simulation in one dimension. The data are consistent with (37).

## 7. Scaling theory of the critical phase

In this section, we derive the scaling exponent for the two-point probability distribution function  $P(m_1, m_2)$  at the critical point  $q = q_c(p)$  in terms of the scaling exponents of the one-point distribution  $P(m)$ . Theoretical analysis and numerical simulations show that the aggregation flux vanishes as  $m \rightarrow \infty$  at the critical point, see figure 3. Therefore, the calculation of the two-point distribution based on constant flux carried out in section 5 does not apply to the critical point. Instead, we will base our argument on the exact result (25) valid just above the critical point and the scaling theory for the EDM developed in [14, 24].

Let us fix the evaporation rate  $p$ . The parameter  $\delta q = q - q_c(p) > 0$  measures the distance to the critical point in the growing phase. We are interested in the limit of  $\delta q \rightarrow +0$  and  $t \rightarrow \infty$ . In this limit  $P(m, \tilde{q}, t)$  displays the scaling form [14, 24]

$$P(m, \delta q, t) \sim \frac{1}{m^{\tau_c}} Y_1 \left( m \delta q^\phi, \frac{m}{t^\alpha} \right), \quad (38)$$



**Figure 5.** Numerical measurement of currents in the exponential phase for the one-dimensional EDM. The aggregation current  $J_{\text{agg}}(m)$  and mass distribution  $P(m)$  decay exponentially with mass in quantitative agreement with mean field theory. Inset: numerical verification of (37) in one dimension.

in terms of three unknown exponents  $\phi$ ,  $\alpha$ ,  $\tau_c$ , and the scaling function of two variables  $Y_1$ . Of interest is one more exponent  $\theta$  defined as follows. In the growing phase, the average mass increases linearly with time, and the mean rate of increase scales with distance to the critical point as  $J \sim \delta q^\theta$ , where  $\theta > 0$  is the growth exponent. In other words,

$$\langle m \rangle \sim \delta q^\theta t, \quad \theta > 0. \quad (39)$$

The growth exponent  $\theta$  is not independent of the exponents  $\tau_c$ ,  $\phi$  and  $\alpha$  which define the scaling of mass distribution: calculating the expected mass  $\langle m \rangle$  using (38) and comparing the answer with (39) we find that [14]

$$\theta = \frac{\phi [1 - \alpha(2 - \tau_c)]}{\alpha}. \quad (40)$$

The exponents  $\alpha$ ,  $\phi$  and  $\tau_c$  are not independent, and are connected through the scaling relation, [25]:

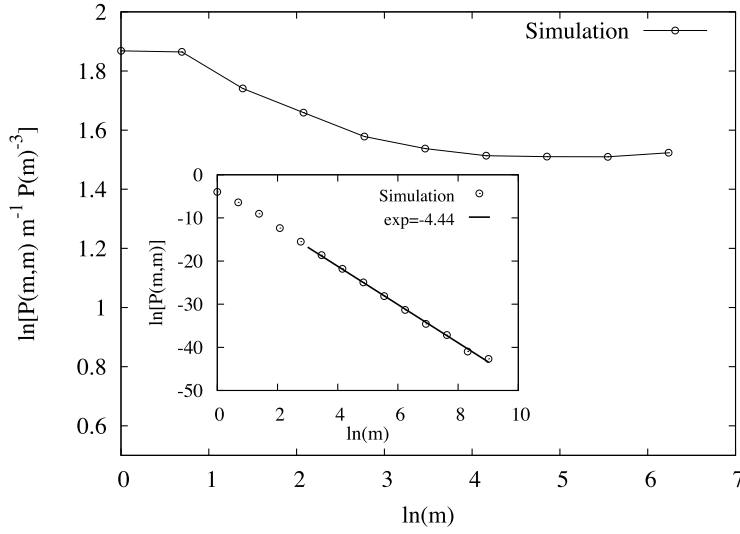
$$\alpha(\tau_c - 1) = \frac{d}{2}. \quad (41)$$

The derivation of (41) is not as straightforward as (40) and is based on a catchment area argument, see [25] for details. We conclude that in the scaling regime, the mass distribution is characterized by two independent scaling exponents, e.g.  $\alpha$  and  $\phi$ .

Now let us consider the two-point probability distribution. In the limit  $t \rightarrow \infty$ , it will have the following scaling form

$$P(m, m, \delta q) \sim \frac{1}{m^{h_c}} Y_2(m \delta q^\phi), \quad (42)$$

where  $Y_2$  is a scaling function of one argument. Note that (42) is based on the assumption that exponent  $\phi$  determines the behaviour of *all* probability distribution functions near



**Figure 6.** Numerical verification of (47) in one dimension: For any  $\tau_c$ , (47) and (38) imply that for  $d = 1$ ,  $P(m, m)/mP(m)^3 \sim m^0$ . Inset:  $P(m, m)$  scales as  $m^{-4.44}$ . This is consistent with  $h_c = \frac{9}{2}$  corresponding to the value of  $\tau_c = \frac{11}{6}$  argued for in [25].

the critical point. In the growing phase, the scaling of  $P(m, m, t)$  as a function of mass  $m$  and flux  $J$  is determined by the constant flux relation:

$$P(m, m, t) \sim \frac{J}{m^h}, \quad m \gg 1, \quad (43)$$

where  $h = 3$ .

If  $Y_2(x) \sim x^{-\mu}$  for  $x \gg 1$ , the comparison of (42) and (43) leads to a pair of equations for exponents  $\mu$  and  $h_c$ :

$$\mu = h - h_c, \quad (44)$$

$$\mu = -\frac{\theta}{\phi}. \quad (45)$$

Solving for  $h_c$  we find that

$$h_c = \frac{1}{d} [d - 2 + \tau_c (2 + d)], \quad (46)$$

which is consistent with the conclusions of [15]. We conclude that at the critical point

$$P(m, m)|_{q=q_c(p)} \sim \left(m^{\frac{1}{d}}\right)^{(2-d)-\tau_c(2+d)}. \quad (47)$$

Note that at the critical point the scaling of the two-point function does depend on  $d$ , the dimensionality of space. This is to be contrasted with the universal scaling law of  $P(m, m, t = \infty)$  in the growing phase due to the constant positive flux of mass in mass space. Note also the independence of  $h_c$  on the evaporation rate  $p$ . In figure 6, we measure  $P(m, m)$  in one dimension at the critical point. The exponent  $\tau_c$  in (47) is unknown, but can be eliminated in terms of the mass distribution  $P(m)$ . The numerical

results in figure 6 confirm the scaling law (47). It is quite surprising and encouraging that the constant flux relation valid in the growing phase combined with a simple heuristic scaling theory allows one to make correct guesses concerning the statistics of the model at the critical point. A rigorous derivation of (47) remains an open problem.

## 8. Summary and conclusions

To summarize, we have studied the phase transition between a growing phase (in which the total mass grows for all time) and an exponential phase (in which the total mass is bounded) in a lattice-based diffusion–aggregation model with injection and evaporation. This transition is controlled by the ratio of the rate of deposition of monomers,  $q$ , and the rate of evaporation,  $p$ . We established a rigorous upper bound on the transition point  $q_c(p)$  valid in all dimensions and showed that the growing phase has an asymptotically constant flux of mass with an associated CFR for the two-point probability distribution function, again valid in all dimensions. From a combination of theoretical analysis and numerical simulations, we conjectured that the growing phase is in the universality class of the aggregation model with deposition but no evaporation, the only effect of evaporation being the modification of the value of the mass flux. Numerical simulations in one dimension show that *all* probability distribution functions of the model in the growing phase scale with mass exactly as they do in the model without evaporation. It would be interesting to verify this theoretically.

The existence of a non-trivial exponential phase, (i.e. the fact that  $q_c(p) > 0$ ) was only established at the level of mean field theory although numerical evidence for the existence of this phase in one dimension is incontrovertible. We note that the presence of the non-equilibrium phase transition can be established analytically in all dimensions [25] in a variant of the model when the diffusion rate is inversely proportional to mass,  $D(m) \sim m^{-1}$ . It would be interesting to see if the analysis carried out in this paper is generalizable to other models of diffusive transport. The exponential phase, if it exists, was shown to be characterized by a global balance between evaporation and deposition *and* a scale-by-scale balance between local fluxes of mass due to evaporation and aggregation. Again, the presence of this scale-by-scale balance has only been established within mean field theory but is strongly supported by numerical simulations in one dimension. Finally, we developed a scaling theory for the critical phase of the model, for which no general analytic tools exist. We applied this theory to predict the scaling of the two-point distribution function and verify the prediction numerically.

In conclusion, the simple aggregation model with deposition and evaporation possesses a wealth of features of considerable interest for non-equilibrium statistical physics in general: the presence of a phase transition (established numerically in one dimension), constant flux relation and anomalous scaling in the growing phase, scale-by-scale balance in the exponential phase, non-trivial scaling behaviour at the critical point. The key to understanding the observed phenomena is an important relationship between conservation laws in the non-equilibrium setting and phase transitions between non-equilibrium steady states which may have more general applicability. Since only some of the above features have been established theoretically, the validation of others, such as the existence of the non-equilibrium phase transition in all dimensions, poses interesting open problems.

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