

## Diffusion-controlled reaction $A + B \rightarrow 0$ in one dimension: The role of particle mobilities and the diffusion-equation approach

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We investigate the influence of different mobilities of  $A$  and  $B$  particles on the time evolution of the particle concentration in the diffusion-controlled reaction  $A + B \rightarrow 0$ . For different concentrations of  $A$  and  $B$  particles, Monte Carlo simulations display two main behaviors, depending on whether the majority species moves or is at rest. An analytical approximation based on the diffusion equation shows reasonable agreement with the Monte Carlo simulations and gives lower bounds for the concentrations' decay.

### I. INTRODUCTION

Diffusion-controlled reactions, where particles move diffusively and react on contact, have been studied intensely during recent years. Although exact mathematical solutions are hard to obtain, several asymptotic formulas for the decay of the particle concentrations have been established by heuristic treatments or by scaling analyses [1–30].

In this paper we focus on the  $A + B \rightarrow 0$  reaction in one dimension. As one already knows from the  $A + B \rightarrow B$  reaction, the asymptotic form depends on which of the particle species is mobile or immobile. Thus, for the one-dimensional target problem ( $A + B \rightarrow B$  with immobile  $A$  and mobile  $B$ ) one obtains for the concentration  $c_A(t)$  of  $A$  particles asymptotically [9]

$$c_A(t) \simeq \exp(-c\sqrt{t}) \quad (1)$$

( $c$  constant), whereas for the trapping problem ( $A$  mobile,  $B$  immobile) one has in one dimension at extremely long times [1,2,7]

$$c_A(t) \simeq \exp(-ct^{1/3}). \quad (2)$$

An analytical treatment of the  $A + B \rightarrow 0$  reaction is more complicated than the  $A + B \rightarrow B$  reaction, due to the fact that in the course of the reaction the number of  $B$  particles also changes; the correlations that develop between the  $A$  and the  $B$  particles are for the  $A + B \rightarrow 0$  reaction more complex than for the  $A + B \rightarrow B$  reaction. Furthermore, as will be evident from the following, the relation between the initial concentrations  $c_A = c_A(0)$  and  $c_B = c_B(0)$  is fundamental for the  $A + B \rightarrow 0$  reaction. Thus one must distinguish between having equal concentrations of particles ( $c_A = c_B$ ) and having one species (say  $A$ ) in the minority ( $c_A < c_B$ ). In the first case ( $c_A = c_B$ ) the decay of concentration follows a  $t^{-1/4}$  law, a fact which by now is well established [3,4,6,15]. The second case ( $c_A < c_B$ ) is more delicate, (see the discussion of Ref. [30]); evidently the time evolution  $c_A(t)$  for  $A + B \rightarrow 0$  is bounded from below by the corresponding expression for

the  $A + B \rightarrow B$  reaction [as given in Eqs. (1) and (2)] and will follow these forms closely if  $c_A \ll c_B$ .

In the present paper we investigate the behavior of the  $A + B \rightarrow 0$  reaction in one dimension: we include cases in which either only one of the species ( $A$  or  $B$ ) moves and also cases in which both species are mobile. In Sec. II we present for all of these cases the results of Monte Carlo simulations of the reaction process on a linear chain. Then, in Sec. III we develop an approximate treatment of the reaction based on diffusion equations. The comparison between the finding of the two approaches and a critical evaluation of the accuracy of the approximation proposed are given in Sec. IV.

### II. MONTE CARLO SIMULATIONS

For the simulation of the  $A + B \rightarrow 0$  reaction in one dimension we start from a random distribution of  $A$  and  $B$  particles; the linear chain (with periodic boundary conditions) consists of  $10^5$ – $10^8$  sites, and each site is either empty or is occupied by one particle only. At each simulation step one of the mobile particles is picked at random and moved to one of its neighboring sites. The time increment for each such step is taken to be the inverse of the total number of mobile particles still present in the system. Thus each mobile particle performs on the average one step per time unit. Note that for the described process the diffusion coefficient of the mobile particles is  $D = \frac{1}{2}$  (in units of lattice distance squared per unit time). Whenever two particles of opposite type meet, they annihilate each other immediately.

Figure 1 shows the time evolution  $c_A(t)$  of the concentration of  $A$  particles. In the simulations two types of conditions were considered: equal initial concentrations,  $c_A = c_B = 0.1$ , and unequal initial concentrations,  $c_A = 0.1$  and  $c_B = 0.2$ . The curves displayed are the average over some ten realizations of each process. Note that the self-averaging is very good, so that the curves in Fig. 1 were not smoothed in any way. The symbols  $A$ ,  $B$ , and  $AB$  indicate the mobile species, respectively.

We note that for equal initial concentrations  $c_A = c_B = 0.1$  the asymptotic regime is reached very early;

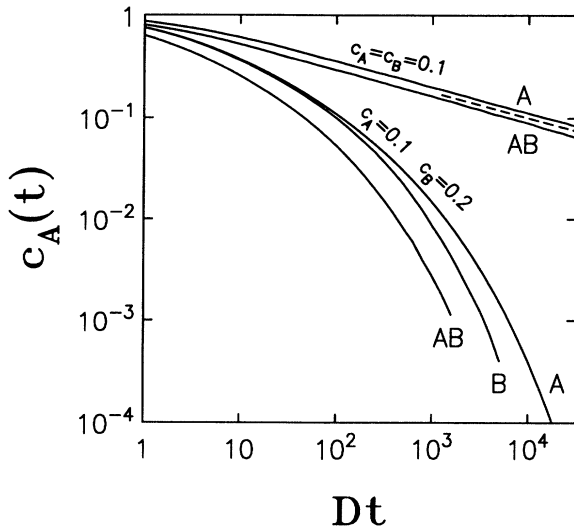


FIG. 1. Time evolution of the concentration  $c_A(t)$  of  $A$  particles for equal ( $c_A=c_B=0.1$ ) and for unequal initial concentrations ( $c_A=0.1$ ,  $c_B=0.2$ ). The curves were obtained from Monte Carlo simulation of the  $A+B\rightarrow 0$  reaction on linear chains of  $10^5$  sites (for  $c_A=c_B$ ) and of  $10^8$  sites (for  $c_A\neq c_B$ ). For  $c_A=c_B$  the curves were averaged over four realizations, for  $c_A\neq c_B$  over ten realizations. The symbols  $A$ ,  $B$ , and  $AB$  indicate the mobile species. The dashed line has the slope  $-\frac{1}{4}$ .

from the figure we infer a slope of  $-0.26$ , very close to the well-known  $t^{-1/4}$  law. For easy reference the dashed line in Fig. 1 indicates the slope  $-\frac{1}{4}$ . Note that the  $t^{-1/4}$  law is obeyed, regardless of whether only one or both types of particles are mobile; for our two cases ( $D_A=D_B$  or  $D_B=0$ ) the two curves differ from each other by a factor of 2 in the time scale. For unequal particle concentrations  $c_A < c_B$ , however, two main behaviors become evident; the decay forms depend on whether the majority species ( $B$ ) is mobile or not. Thus, the situation in which both  $A$  and  $B$  move leads to a similar decay as that obtained when only  $B$  (the majority species) is mobile; for  $D_A=D_B$  and  $D_A=0$  the corresponding decay laws differ only by a factor of 2 in the time scale. However, very different temporal behavior obtains when only the minority species ( $A$ ) is mobile, the decay being considerably slower than in the former situation. Note that this feature parallels the findings for the  $A+B\rightarrow B$  reaction, where the decay due to trapping, Eq. (2), is at long times slower than that of the target problem, Eq. (1).

### III. THE DIFFUSION-EQUATION APPROACH

In a continuous-medium representation a reaction between two diffusing species can be described by the two differential equations

$$\frac{\partial c_A(x,t)}{\partial t} = D_A \Delta c_A(x,t) - R(x,t)$$

and

$$\frac{\partial c_B(x,t)}{\partial t} = D_B \Delta c_B(x,t) - R(x,t), \quad (3)$$

where  $c_A(x,t)$  and  $c_B(x,t)$  are the local concentrations,  $D_A$  and  $D_B$  are the corresponding diffusion coefficients, and  $R(x,t)$  is a term that describes the local decrease in the particle concentrations due to the reaction. In general,  $R(x,t)$  depends on the joint probability density  $\rho(r,t)$  for finding particles  $A$  and  $B$  at time  $t$  at the mutual distance  $r$ . In general,  $R(x,t)$  is not expressible as a product of the local concentrations  $c_A(x,t)$  and  $c_B(x,t)$ ; see, for instance, Ref. [16]. However, in Eq. (3) it is only important that the particles react pairwise, so that the same expression for  $R(x,t)$  describes both the decrease in the number of  $A$  and in the number of  $B$  particles.

#### A. Equal diffusion coefficients $D_A=D_B$

In the case of equal diffusion coefficients  $D_A=D_B\equiv D$ , Eqs. (3) lead to a simple diffusion equation for the variable  $q(x,t)=c_A(x,t)-c_B(x,t)$  [3,4]:

$$\frac{\partial q(x,t)}{\partial t} = D \Delta q(x,t). \quad (4)$$

The case of unequal diffusion coefficients  $D_A\neq D_B$  will be considered in the next subsection. We note that Eq. (4) is a problem that (for a given set of initial conditions) can be solved by simple analytical means. Here we proceed by displaying this solution; in a second step we will use the results to obtain approximate forms for  $c_A(t)$ .

Let  $q(x,0)$  be the initial distribution of  $q$ . Then  $q(x,t)$  is given by the equation

$$q(x,t) = \int q(\xi,0) G(x-\xi,t) d\xi, \quad (5)$$

where

$$G(x,t) = (4\pi Dt)^{-1/2} \exp\left[-\frac{x^2}{4Dt}\right] \quad (6)$$

is the Green's function of the diffusion equation, Eq. (4). If  $q(x,0)$  is random, has finite dispersion and no long-range spatial correlations (that is true for Poissonian, i.e., completely random initial distribution of particles), the distribution of  $q(x,t)$  will approach a Gaussian for large  $t$ ; this is so because for large  $t$   $G(x,t)$  is rather broad and the integral on the right-hand side of Eq. (5) can be treated as a sum of a large number of independent terms, for which the conditions of the central limit theorem apply [29]. In fact, a recent numerical analysis of the  $q(x,t)$  behavior shows that in  $d=1$  the distribution is very well represented by a Gaussian for times such that  $Dt > 100$  [31].

In the Gaussian regime the distribution of  $q$  is fully characterized by the mean  $\mu = \langle q \rangle$  and the dispersion  $\sigma^2 = \langle q^2 \rangle - \langle q \rangle^2$ ; here  $\langle \rangle$  denotes the ensemble average. Averaging Eq. (5) gives

$$\langle q(x,t) \rangle = \int G(x-\xi,t) \langle q(\xi,0) \rangle d\xi, \quad (7)$$

because the Green's function is nonrandom. Thus, since  $\langle q(\xi,0) \rangle = c_A - c_B$ , we have  $\mu = \langle q(x,t) \rangle = c_A - c_B$ . The time evolution of the second moment  $\langle q^2 \rangle$  is given by the equation

$$\begin{aligned} \langle q^2(t) \rangle &\equiv \langle q(x,t)q(x,t) \rangle \\ &= \int \int G(\xi,t)g(\eta-\xi)G(-\eta,t)d\xi d\eta, \end{aligned} \quad (8)$$

where  $g(x) \equiv \langle q(y,0)q(y+x,0) \rangle$ . In terms of the spatial Fourier transform of  $g(x)$  and  $G(x,t)$ , say  $g(k) = \int g(x)e^{ikx}dx$ , the right-hand side of Eq. (8) can be written as

$$\langle q^2(t) \rangle = \frac{1}{2\pi} \int g(k)\exp(-2Dtk^2)dk, \quad (9)$$

where the explicit form of the Fourier transform of Eq. (6),  $G(k,t) = \exp(-Dtk^2)$ , was used.

In the case of an initially totally disordered distribution of point particles one finds (see the Appendix)

$$g(k) = c_A + c_B + 2\pi(c_A - c_B)^2\delta(k). \quad (10)$$

Substituting this result into Eq. (9), one obtains the second moment  $\langle q^2(t) \rangle$  and hence the dispersion  $\sigma^2(t)$ :

$$\sigma^2(t) = \frac{c_A + c_B}{\sqrt{8\pi Dt}}. \quad (11)$$

At longer times, for which the  $q$  distribution is Gaussian, one has as probability density  $P(q,t)$

$$\begin{aligned} P(q,t)dq &= \frac{1}{\sqrt{2\pi\sigma^2(t)}} \exp\left[-\frac{(q-\mu)^2}{2\sigma^2(t)}\right]dq \\ &= \left[\frac{2Dt}{\pi}\right]^{1/4} \sqrt{c_A + c_B} \\ &\quad \times \exp\left[-\frac{(q - c_A + c_B)^2\sqrt{2\pi Dt}}{c_A + c_B}\right]dq. \end{aligned} \quad (12)$$

In order to calculate the average particle concentration  $c_A(t) = \langle c_A(x,t) \rangle$  from  $P(q,t)$  we must use an approximation. The main idea of this approximation is to assume that in regions where the  $A$  particles are in the majority there are no  $B$  particles present and vice versa. This situation is approximately fulfilled when the local reaction rate (given by  $R$ ) is large compared to the diffusion term, so that one has clear-cut  $A$  and  $B$  regions. Then one can identify  $q(x,t)$  with  $c_A(x,t)$  when  $q(x,t)$  is positive, and with  $c_B(x,t)$  otherwise. Generally we define

$$\bar{c}_A(x,t) = q(x,t)\Theta(q(x,t)) \quad (13)$$

and

$$\bar{c}_B(x,t) = -q(x,t)\Theta(-q(x,t)),$$

where  $\Theta(x) = 0$  for  $x$  negative and  $\Theta(x) = 1$  for  $x$  positive. In the following we use  $\bar{c}_i(x,t)$  as an approximation to  $c_i(x,t)$ , where  $i$  is  $A$  or  $B$ . Note that one always has  $\bar{c}_i(x,t) \leq c_i(x,t)$  for both  $i = A$  and  $i = B$ , i.e., our approximation *underestimates* the true particle concentrations. As an example, our approximation gives for the average concentration of  $A$  particles

$$\bar{c}_A(t) = \langle q(x,t)\Theta[q(x,t)] \rangle = \int_0^\infty q P(q,t)dq. \quad (14)$$

For the probability density  $P(q,t)$  given by Eq. (12), one can evaluate Eq. (14) readily, obtaining

$$\bar{c}_A(t) = \frac{\sigma(t)}{\sqrt{2\pi}} \exp\left[-\frac{\mu^2}{2\sigma^2(t)}\right] + \frac{\mu}{2} \operatorname{erfc}\left[-\frac{\mu}{\sqrt{2\sigma(t)}}\right], \quad (15)$$

where  $\operatorname{erfc}(x)$  is the complementary error function [32]. For equal initial concentrations  $c_A = c_B$ , the mean  $\mu$  equals zero and  $\bar{c}_A(t)$  simplifies to

$$\bar{c}_A(t) = \frac{\sigma(t)}{\sqrt{2\pi}} = \frac{(c_A + c_B)^{1/2}}{2^{5/4}\pi^{3/4}}(Dt)^{-1/4}. \quad (16)$$

Notice that this result reproduces the  $t^{-d/4}$  law.

On the other hand, in the case that the  $A$  particles are in the minority,  $c_A < c_B$ , one obtains from Eq. (15) at large  $t$

$$\bar{c}_A(t) \simeq t^{-3/4} \exp\left[-\frac{(c_A - c_B)^2}{(c_A + c_B)}\sqrt{2\pi Dt}\right]. \quad (17)$$

Note that the results displayed in Eqs. (15), (16), and (17) have exactly the same form as those presented in Ref. [30] (see Eqs. (15), (16), and (20) of Ref. [30], where one sets  $w = c_A + c_B$ ,  $\Delta = c_B - c_A$ , and  $N = (8\pi Dt)^{1/2}$ ). This is quite remarkable, since the results in Ref. [30] were derived based on a method using fluctuation statistics. Furthermore, in Ref. [30] we pointed out that in the case in which only the minority species ( $A$ ) moves and the majority species is at rest, Eq. (15) is only approximate, being valid in an intermediate time regime only and not for large  $t$ .

## B. Unequal diffusion coefficients $D_A \neq D_B$

For unequal diffusion coefficients  $D_A \neq D_B$  we cannot proceed as in Eq. (4), since from Eqs. (3) one now obtains

$$\frac{\partial q(x,t)}{\partial t} = D_A \Delta c_A(x,t) - D_B \Delta c_B(x,t), \quad (18)$$

which is not a closed equation for  $q$ . A way out of this difficulty is to invoke again the picture of well-separated  $A$  and  $B$  domains, and approximate  $c_i(x,t)$  by  $\bar{c}_i(x,t)$  according to Eq. (13). Formally one can now write

$$\frac{\partial q(x,t)}{\partial t} = \nabla D(q) \nabla q(x,t), \quad (19)$$

where

$$D(q) = \begin{cases} D_A & \text{if } q > 0 \\ D_B & \text{if } q < 0. \end{cases} \quad (20)$$

Note that care has to be exercised with Eq. (19) in the case when  $q \equiv 0$ . Furthermore, even in this form Eq. (19) cannot be treated readily analytically. We circumvent these problems by reverting to a numerical procedure and consider the discrete analog of Eq. (19); for a linear chain (with periodic boundary conditions) one has

$$\frac{\partial}{\partial t} q_i = q_{i+1}D(q_{i+1}) - 2q_iD(q_i) + q_{i-1}D(q_{i-1}), \quad (21)$$

where (if  $q_i$  is positive)  $q_i$  is taken to be the probability for finding an  $A$  particle on site  $i$ ; if  $q_i$  is negative,  $-q_i$  is

the corresponding probability for the  $B$  particles.

Equation (21) has the form of a master equation; the change in the concentration of particles at site  $i$  is due to particle transfers between neighboring sites, the transition rates from one site to the other depending on the particle type (i.e.,  $q > 0$  or  $q < 0$ ).

We solved Eq. (21) numerically for initial conditions corresponding to the Monte Carlo simulations, i.e.,  $q_i = 1$  with probability  $c_A(0)$  and  $q_i = -1$  with probability  $c_B(0)$ . Again we considered the three cases: (i)  $D_A = D$ ,  $D_B = 0$ ; (ii)  $D_A = 0$ ,  $D_B = D$ ; and (iii)  $D_A = D_B = D$ . For the discretization of Eq. (21) in time we chose time intervals such that  $D\Delta t = \frac{1}{4}$ , as it is convenient for solving diffusion equations.

First we want to illustrate the difference between the Monte Carlo process described in Sec. II and the diffusion approximation described by Eq. (21). Whereas in the Monte Carlo simulation the particles keep their discrete nature, i.e.,  $q_i = \pm 1$  if site  $i$  is occupied by an  $A$  or  $B$  particle and  $q_i = 0$  if site  $i$  is empty, in the diffusion approximation  $q_i$  can take any value between  $-1$  and  $1$ , depending on the probability of finding an  $A$  or a  $B$  particle on site  $i$ .

Starting with the same initial distribution of particles on a linear chain of 1280 sites, we display in Fig. 2 the particle distribution  $q_i$  at times corresponding to  $Dt = 128$ , the distributions resulting from the Monte Carlo process on the one hand and from the numerical solution of Eq. (21) on the other hand. Depending on which of the particle species is mobile (only  $A$  or only  $B$  or  $A$  and  $B$ ), the particle distributions are different. In each case the first picture in Fig. 2 indicates the particle distribution resulting from the Monte Carlo process, whereas the second picture displays the solution of Eq. (21). Each site  $i$  of the lattice is represented by a vertical line whose length corresponds to the value of  $q_i$ .

In the cases where only one particle species ( $A$  or  $B$ ) is

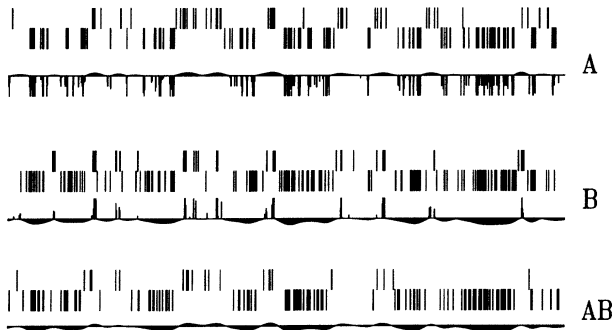


FIG. 2. Particle distributions along the line at times corresponding to  $Dt = 128$ . Three cases are considered: mobile species  $A$ ,  $B$ , and  $AB$ . In each case the upper picture gives the Monte Carlo results, where the lines drawn upwards (downwards) denote the  $A$  ( $B$ ) particles, respectively. The lower picture in each case represents the numerical solution for  $q$ , where probabilities for mobile species are smeared out. The initial distribution of particles is identical in all cases.

mobile, the positions of the mobile particles are smeared out due to diffusion, which results in a rather smooth behavior of  $q$  in the regions where no immobile particles have survived. The positions of the immobile particles, however, remain unchanged, but their survival probability decreases according to the flux of mobile particles.

Qualitatively, in the regions where the solution of Eq. (21) predicts a positive (negative) value for  $q$ , one actually finds clusters of  $A$  ( $B$ ) particles also in the Monte Carlo simulations. In some cases, however, one also finds an  $A$  ( $B$ ) particle in a region, where  $q$  is negative (positive). This is due to the fact that  $q$  is not the real probability for finding a particle, but it is an approximation for the difference between the probabilities for finding an  $A$  or a  $B$  particle. Thus, the summation of all positive values of  $q_i$  in the case of the diffusion approximation [Eq. (21)] does not coincide with the exact number of  $A$  particles present in the Monte Carlo process. As already stated above, this procedure always underestimates the real concentration of  $A$  particles.

#### IV. DISCUSSION AND CONCLUSIONS

Figure 3 displays a comparison between the results for  $c_A(t)$  obtained via direct Monte Carlo simulations of the reaction and via the solution of Eq. (21). The parameters are the same as in Fig. 1. As expected, the curves resulting from the diffusion approximation approach the actual concentration of  $A$  particles found in Monte Carlo simulations from below, as the diffusion approximation underestimates the concentration of particles. The relative error, however, is smaller than 25% in all cases considered. The approximation works very well in the cases of equal concentrations ( $c_A = c_B$ ) and in the case when only the minority species ( $A$ ) is mobile. We note that in

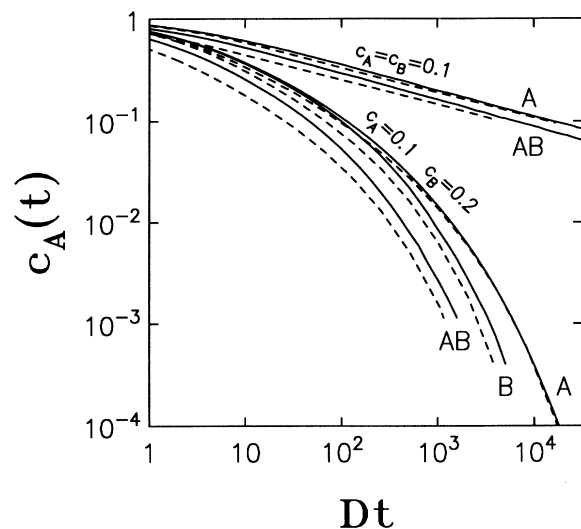


FIG. 3. The solid lines give the results of the Monte Carlo simulations and correspond to the data of Fig. 1; here the dashed lines indicate the solutions of Eq. (21) (see main text for details).

the case where  $A$  and  $B$  are mobile, the concentration decay resulting from the numerical solution of Eq. (21) coincides with the analytical result given in Eq. (15).

In summary, we have shown that Monte Carlo simulations of the diffusion-controlled reaction  $A + B \rightarrow 0$  in one dimension tend to indicate that in the case of unequal concentrations ( $c_A < c_B$ ) of  $A$  and  $B$  particles, there are two different universality classes insofar as the decay of concentration of  $A$  particles is concerned. The cases in which only the majority species or both species are mobile show the same temporal behavior, the only difference being a factor of two in the time scale. The case in which only the minority species ( $A$ ) is mobile, however, results in a quite different temporal behavior. These findings parallel the results for the trapping and the target problem.

An approximate treatment of the  $A + B \rightarrow 0$  reaction, which is based on the diffusion equation, displays qualitatively the same behavior as the Monte Carlo simulations, the relative error being less than 25%. In the case in which both particle species are mobile the approximation allows us to derive an approximate analytical solution for the average concentration  $c_A(t)$  of the  $A$  particles.

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#### APPENDIX

Let us consider a random distribution of point particles on a line segment consisting of cells. We take  $N_A$  and  $N_B$  to be the total number of  $A$  and  $B$  particles, with  $N_A \ll L$  and  $N_B \ll L$ . Under these circumstances each cell is occupied at most by one particle. Consider now the discrete correlation function  $\langle q_i q_j \rangle$  corresponding to  $g(x, y) = \langle q(x) q(y) \rangle$  of the main text. One has now  $q_i = 1$  if the  $i$ th cell is occupied by an  $A$  particle,  $q_i = -1$  if the  $i$ th cell is occupied by a  $B$  particle, and  $q = 0$  elsewhere. For randomly distributed  $A$  and  $B$  particles one has evidently for  $i \neq j$

$$g(i, j) = \langle q_i q_j \rangle = \langle q_i \rangle \langle q_j \rangle = \langle q_i \rangle^2 \\ = \left[ \frac{N_A - N_B}{L} \right]^2 = (c_A - c_B)^2, \quad (\text{A1})$$

where we used the relations  $c_A = N_A/L$  and  $c_B = N_B/L$ . On the other hand, for  $i = j$  it follows that

$$g(i, i) = \langle q_i^2 \rangle = \frac{N_A + N_B}{L} = c_A + c_B. \quad (\text{A2})$$

Reverting to the continuum description it follows that

$$g(x, y) = (c_A + c_B) \delta(x - y) + (c_A - c_B)^2. \quad (\text{A3})$$

Thus  $g(x, y)$  depends only on the difference  $x - y$ :  $g(x, y) = g(x - y)$ . Fourier transforming Eq. (A3) leads to Eq. (10) of the main text.

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