Solution of the static pair annihilation process

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The pair annihilation of identical static particles initially distributed at random on a one-dimensional lattice is studied for exponential and power-law interactions. A shielding approximation is introduced to solve the hierarchy of equations describing the process, and the density and pair correlation of surviving particles are calculated. The analytical and numerical results of this approximation are in good agreement with Monte Carlo simulations, showing that the approximation correctly describes the long-time regime in all cases. [S1063-651X(97)07706-4]

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I. INTRODUCTION

In the static annihilation model, a set of A particles are randomly distributed on a lattice and removed by a fusion reaction $A+A\to 0$, with isotropic reaction rate $w(r)$, for any pair of particles separated by distance r. Extensive studies (see [1] and references therein) have shown that the process is dominated at time t by fluctuations of the number of particles in a volume of size roughly determined by the reaction radius [2] $R(t)$ given by $tw(R(t))\approx 1$. The main observables are the density of surviving particles $\rho(t)$ and the two particle correlation function $g(r,t)$. In the one-dimensional case, it appears that the density decays as $\rho(t)=C/R(t)$, while the correlation ultimately scales as $g(r,t)=g_\infty(z=r/R(t))$.

In an earlier paper [3], we gave analytical expressions for the constant C and the asymptotic form of $g_\infty(z)$ for a tunneling law interaction $w(r)=w_0\exp(-r/r_0)$ such as might be found for annihilation of localized triplet electronic states of aromatic molecules in rigid solution [4,5]. We introduced a staggered annihilation model, in which the reaction occurs by stages corresponding to pairs of particles separated by $1,2,3,\ldots,N$ lattice units. This is intuitively justified for small r_0 compared to the lattice spacing and leads to a density decaying asymptotically as $e^{-\gamma}/2N$, where γ is Euler's constant. We argued that the true static annihilation model is recovered by the identification $N=R(t)$, giving $C=e^{-\gamma}/2$, in agreement with the density and pair correlation in Monte Carlo simulations of the true static annihilation process.

Now this implies that C and $g_\infty(z)$ might be independent of the form of $w(r)$, insofar as the static and the staggered static processes may be identified. Our purpose here is to investigate this. We therefore solve the static annihilation model without the staggered interaction approximation, in the spirit of [7], by using a Kirkwood-like uncoupling approximation to close the hierarchy of equations of the static model. We then recover all the analytical results suggested by the staggered reaction model for the exponential case. However, we find that for power-law interactions $w(r)=w_0/r^\alpha$, $\alpha>1$, the density $\rho(t)$ behaves asymptotically like $C_\alpha/R(t)$, where $R(t)=t^{1/\alpha}$ and the constant C_α depends on α. The asymptotic pair correlation also depends on α. Both quantities tend to their values for the exponential reaction as α tends to infinity. The results are again in very good agreement with the Monte Carlo simulation of the exact process. Section II below is devoted to the hierarchy of equations defining the static annihilation process on a one-dimensional lattice, using a shielding approximation, which is to assume that two particles do not react while there are still other particles between them. We expect this to be an accurate approximation for reaction rates decaying sufficiently steeply with r and especially for dilute systems. We have performed Monte Carlo simulations to check this and conclude that the approximation correctly produces the asymptotic form of the decay for all interactions studied here. This approximation closes the hierarchy of equations for the density and the correlation function, opening the way to an exact solution of the model in the asymptotic limit. Section III describes the analytical solution for the exponential interaction, recovering our earlier results [3] in a much more direct way. Finally, Sec. IV applies the same method to the power-law interaction. Numerical results are given for $2\leqslant\alpha\leqslant 6$. Although the constant C_α is close to its value for the exponential interaction $e^{-\gamma}/2$, it shows some dependence on α. The departure from the exponential case is more pronounced for the pair correlation, which indicates that the self-ordering tendency decreases with α. The correctness of the results is checked by a comparison with Monte Carlo simulations of the true static annihilation model.

II. HIERARCHICAL EQUATIONS WITHIN THE SHIELDING APPROXIMATION

The well-known hierarchy [6] used to describe the dynamics of annihilation begins with the evolution of the density

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$$-\frac{d}{dt}\rho(t)=2\sum_{r=1}^{\infty} w(r)P_2(r,t), \tag{1}$$

where $P_2(r,t)$ is the probability (concentration per pair of lattice sites) of pairs of particles A_1A_2 with separation r at time t. The separation is measured in lattice spacings and the sum runs over half the lattice, the factor 2 arising from symmetry. The evolution of $P_2(r,t)$ is governed by two kinds of events: the pair A_1A_2 may react or one of its members may react with a third particle A_3. When A_3 is outside the interval $[A_1,A_2]$, the shielding approximation implies that only the nearer member of the pair reacts with A_3. Thus

$$-\frac{d}{dt}P_2(r,t)=w(r)P_2(r,t)+2\sum_{r'<r} w(r')P_3(r',r-r',t)$$
$$+2\sum_{r'} w(r')P_3(r',r,t), \tag{2}$$

where $P_3(r_1,r_2,t)$ is the probability of finding three successive particles $A_1A_2A_3$ with separations $A_1A_2=r_1$ and $A_2A_3=r_2$ and we have used the symmetry $P_3(r_1,r_2,t)=P_3(r_2,r_1,t)$. The generalization of these equations is straightforward. Let $P_n(\{r_i\},t)$ be the probability for n successive particles $A_1A_2\cdots A_n$ with separations $A_iA_{i+1}=r_i$, $1\leqslant i\leqslant n-1$. Then

$$-\frac{d}{dt}P_n(\{r_i\},t)$$
$$=\sum_{j=1}^{n-1} w(r_j)P_n(\{r_i\},t)+2\sum_{j=1}^{n-1}\sum_{r_{j'}<r_j} w(r_{j'})P_{n+1}$$
$$\times(r_1,\ldots,r_{j-1},r_{j'},r_j-r_{j'},r_{j+1},\ldots,r_{n-1},t)$$
$$+2\sum_{r'} w(r')P_{n+1}(\{r_i\},r',t). \tag{3}$$

The right-hand side of Eq. (3) describes in order the following: annihilations A_j-A_{j+1}, reactions of A_j or A_{j+1} with some $A_{j'}$ lying between them, and annihilation of A_1 or A_n with particles outside the interval $[A_1,A_n]$. It can be shown that Eq. (3) is solved by a factorized probability

$$P_n(\{r_i\},t)=\rho(t)\prod_{j=1}^{n-1}\gamma(r_j,t), \quad n\geqslant 2 \tag{4}$$

as long as the density and the function $\gamma(r,t)$ satisfy Eqs. (2) and (3). In terms of these variables, Eqs. (2) and (3) can be expressed as

$$-\frac{d}{dt}\rho(t)=2\rho(t)\sum_{r} w(r)\gamma(r,t), \tag{5}$$

$$-\frac{d}{dt}\gamma(r,t)=w(r)\gamma(r,t)+2\sum_{r'<r} w(r')\gamma(r',t)\gamma(r-r',t). \tag{6}$$

The initial conditions are $\rho(t=0)=\rho_0$ and $\gamma(r,t=0)=\rho_0$ because for an initially random distribution $P_2(r,t=0)=\rho_0\gamma(r,t=0)=\rho_0^2$. Thus, from Eq. (5),

$$\rho(t)=\rho_0\exp\left(-2\sum_{r} w(r)\int_0^t\gamma(r,t')dt'\right). \tag{7}$$

On the other hand, the pair correlation function $g(r,t)$, normalized to $g(r=\infty,t)=1$ for all times, is defined via $P_2(r,t)=\rho^2(t)g(r,t)$ or

$$g(r,t)=\gamma(r,t)/\rho(t), \tag{8}$$

so that we can calculate the density and the pair correlation if we can solve Eq. (6) for $\gamma(r,t)$. It can be shown by induction that $\gamma(r,t)$ is of the form

$$\gamma(r,t)=A(r)e^{-w(r)t}+\sum_{i=1}^{I(r)} A(r,i)e^{-\mathcal{L}_i(w)t}, \tag{9}$$

where the summation is over all integer partitions of r and the coefficients $A(r)$ and $A(r,i)$, which are step by step computable, are functions of ρ_0 and $w(r)$. For example, for $r=4$, there are four partitions $[I(r)=4]$, namely, (3,1),(2,2),(2,1,1),(1,1,1,1), corresponding to $i=1,\ldots,4$. The corresponding values of $\mathcal{L}_i(w)$ are $\mathcal{L}_1(w)=w(1)+w(3)$, $\mathcal{L}_2(w)=w(2)+w(2)=2w(2)$, $\mathcal{L}_3(w)=w(2)+2w(1)$, and $\mathcal{L}_4(w)=4w(1)$. The general form is $\mathcal{L}_i(w)=\Sigma_j w(j)$, where j runs over the ith partition. We mention form (9) because it determines the beginning of the annihilation process, but our interest below is the long-time behavior, where we shall look for a scaling form of $\gamma(r,t)$:

$$\gamma(r,t)=h(z)\frac{1}{R(t)} \quad \text{with} \quad z=\frac{r}{R(t)}, \tag{10}$$

where $R(t)$ is the ‘‘reaction radius’’ defined by $tw(R(t))=1$. Physically, in cases with steep radial decay of $w(r)$, annihilation processes with r greater than $R(t)$ at a given time are unlikely. Our aim in Sec. III is to find $h(z)$, if it exists, thus determining the asymptotic behavior of the density and the correlation function, since from Eq. (8),

$$g(r\to\infty,t)=1=\frac{\gamma(r=\infty,t)}{\rho(t)}=\frac{h(\infty)}{R(t)\rho(t)}.$$

Thus

$$\lim_{t=\infty}\rho(t)=\frac{h(\infty)}{R(t)} \tag{11}$$

and

$$g(r,t)=\frac{\gamma(r,t)}{\rho(t)}\simeq\frac{h(z)R(t)}{R(t)h(\infty)}=\frac{h(z)}{h(\infty)},$$

so

$$\lim_{t=\infty}g(r,t)=g_\infty(z)=\frac{h(z)}{h(\infty)}. \tag{12}$$

The determination of $h(z)$, which appears to be rate dependent, is deferred to Sec. III, but we prepare for the derivation by examining here a general constraint implied by Eq. (6) and the initial conditions. On introducing the generating functions $\Gamma(x,t)=\Sigma_r x^r \gamma(r,t)$ and $\Gamma_w(x,t)=\Sigma_r x^r w(r)\gamma(r,t)$, Eq. (6) is equivalent to

$$\frac{-d}{dt}\Gamma(x,t)=[1+2\Gamma(x,t)]\Gamma_w(x,t), \tag{13}$$

which can be integrated to yield

$$1+2\Gamma(x,t)=[1+2\Gamma(x,0)]\exp\left(-2\int_0^t \Gamma_w(x,t')dt'\right). \tag{14}$$

From the initial conditions, $1+2\Gamma(x,0)=1+2\rho_0 x/(1-x)$, which can be written as $\exp(2\Sigma_r x^r \omega_r)$, with $\omega_r=\{1-(1-2\rho_0)^r\}/2r$. Thus Eq. (14) becomes

$$1+2\Gamma(x,t)=\exp\left[2\sum_r x^r\left(\omega^r - w(r)\int_0^t \gamma(r,t')dt'\right)\right], \tag{15}$$

which will be used below.

We should like to conclude this section by justifying the shielding approximation on the basis of numerical experiments involving several hundred thousand events per case studied. Simulations of the $A+A\rightarrow 0$ process were performed for A particles on a line, as described in detail in [3]. Both the exact and the shielded interaction were simulated. Slight differences in the density and the pair correlation for the exact and the shielded interactions quickly drop below the level of statistical significance as the reaction radius grows. Figure 1 compares the density and the correlation functions for the exact and the shielded dynamics with a dipolar interaction $w(r)=1/r^6$ (we set $w_0=1$ throughout the following).

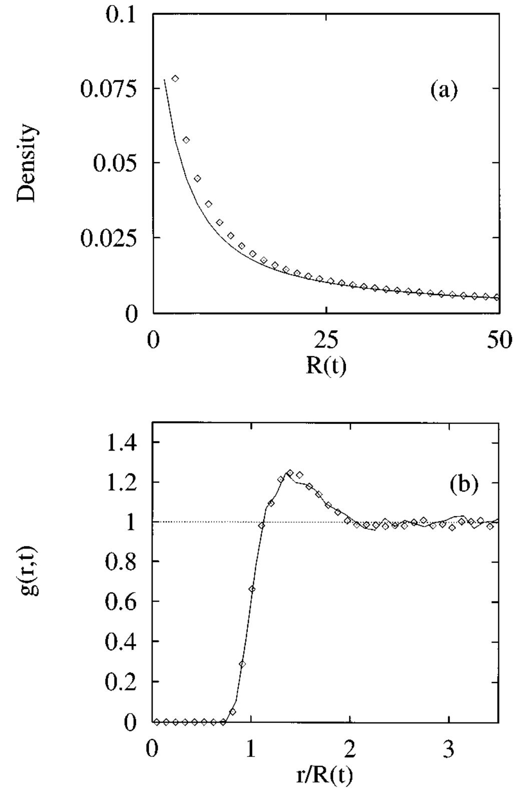

FIG. 1. Comparison of (a) the densities and (b) the asymptotic pair correlation in the $A+A\rightarrow 0$ reaction with $w(r)=1/r^6$, computed by the Monte Carlo simulation of the exact process (curves) and the shielding approximation (diamonds).

III. STATIC ANNIHILATION BY A TUNNELING INTERACTION

Setting unimportant constants to unity, we consider the case $w(r)=e^{-r}$, for which $R(t)=\ln(t)$ and look for the scaling form of solution of Eq. (6),

$$\gamma(r,t)=\frac{h(z)}{\ln(t)}, \quad z=\frac{r}{\ln(t)}. \tag{16}$$

On replacing the summation over r' in Eq. (6) and changing variables to $z'=r'/\ln(t)$, we have

$$\frac{d}{dz}[zh(z)]=t\ln t\left[\frac{h(z)}{t^z}+2\int_0^z dz'\,\frac{h(z')h(z-z')}{t^{z'}}\right]. \tag{17}$$

Thus, as t goes to infinity,

$$h(z)=0 \quad \text{for } 0\leqslant z\leqslant 1 \tag{18}$$

and $h(z)$ for larger z is defined piecemeal on intervals of unit length, the effective range of integration in Eq. (17) being $[1,z-1]$ instead of $[0,z]$. The integral does not contribute for $1<z\leqslant 2$ and

$$h(z)=\frac{h(1)}{z} \quad \text{for } 1<z\leqslant 2, \tag{19}$$

where the constant of integration $h(1)$ will be determined later. For $2\leqslant z\leqslant 3$, retaining the leading term in Eq. (17) yields

$$\frac{d}{dz}[zh(z)]=2h^2(1)t\ln t\int_1^{z-1}\frac{dz' t^{-z'}}{z'(z-z')}\sim\frac{2h^2(1)}{(z-1)} \tag{20}$$

and

$$h(z)=\frac{h(1)}{z}[1+2h(1)\ln(z-1)] \quad \text{for } 2\leqslant z\leqslant 3, \tag{21}$$

where the constant of integration is fixed so that $h(z)$ is continuous at $z=2$. The solution for larger z involves complicated integrals and it is simpler to use the constraints (15) to find $h(1)$ and $h(\infty)$. Relation (18) means that for large t, $\gamma(r,t)=0$ for $r\leqslant R(t)$, in line with the phenomenological interpretation of $R(t)$ as the minimal distance between surviving particles. But this implies that $1+2\Gamma(x,t)=1+O(x^{R(t)})$ and thus all terms in x^r for $1\leqslant r\leqslant R(t)$ on the right-hand side of Eq. (15) must vanish, i.e.,

$$w(r)\int_0^t \gamma(r,t')dt'=\omega^r \quad \text{for } r\leqslant R(t), \tag{22}$$

which in turn leads to

$$2\sum_{r=1}^{R(t)} w(r)\int_0^t \gamma(r,t')dt' = \sum_{r=1}^{R(t)} \frac{1-(1-2\rho_0)^r}{r} \sim \gamma + \ln[2\rho_0 R(t)] \qquad (23)$$

for large t. In this equation, $\gamma = 0.577\ldots$ is Euler's constant. Inserting Eq. (23) into the density, we obtain

$$\rho(t) = \frac{e^{-\gamma}}{2\ln(t)}\rho_w(t), \qquad (24)$$

with

$$\rho_w(t) = \exp\left(-2\sum_{r>R(t)} w(r)\int_0^t \gamma(r,t')dt'\right). \qquad (25)$$

In expression (25), $\gamma(r,t') = \rho(t')g(r,t') \sim \rho(t')$ under the reasonable assumption that outside the reaction radius the correlation is of order 1, as in the initial state. Since $\Sigma_{r>R(t)} w(r) = 1/t$, we obtain

$$\rho_w(t) \sim \exp\left(-2/t\int_0^t \rho(t')dt'\right) \sim 1$$

and the final result is

$$\lim_{t\to\infty}\rho(t) = \frac{e^{-\gamma}}{2\ln(t)}, \qquad (26)$$

which by comparison with Eq. (11) gives directly $h(\infty) = e^{-\gamma}/2$.

In order to determine $h(1)$, we again turn to Eq. (15), which yields

$$\gamma(r,t) = \omega_r - w(r)\int_0^t \gamma(r,t')dt' \quad \text{for } R(t) < r \leq 2R(t).$$

For large r, γ reduces to $\gamma(r,t) = w_r \simeq 1/2r$, i.e., $h(z) = 1/2z$ for $1 < z \leq 2$, in agreement with Eq. (19), whence $h(1) = 1/2$. On collecting these results and using Eq. (12) to compute $g_\infty(z)$, we find from Eqs. (18), (19), and (21) that

$$g_\infty(z) = \begin{cases} 0, & 0 \leq z \leq 1 \\ \dfrac{e^\gamma}{z}, & 1 < z \leq 2 \\ \dfrac{e^\gamma}{z}[1+\ln(z-1)], & 2 \leq z \leq 3. \end{cases} \qquad (27)$$

Results (26) and (27) are in complete agreement with those obtained in [3] by mapping the static annihilation problem onto a staggered annihilation model. We know from this previous work that these expressions agree with Monte Carlo simulations and can be used over a wide range of time.

IV. STATIC ANNIHILATION WITH A POWER-LAW INTERACTION

Consider now the case $w(r) = r^{-\alpha}$. Let us look for solutions of Eq. (6) of the scaling form

$$\gamma(r,t) = h(z)t^{-1/\alpha}, \quad z = rt^{-1/\alpha}, \qquad (28)$$

in agreement with Eq. (10) and our definition $R(t) = t^{1/\alpha}$. On inserting expression (28) into Eq. (6) and dividing throughout by $t^{-1-(1/\alpha)}$, one finds the time-independent equation for $h(z)$,

$$\frac{d}{dz}[zh(z)] = \frac{\alpha h(z)}{z^\alpha} + 2\alpha\int_0^z \frac{dx}{x^\alpha}h(x)h(z-x). \qquad (29)$$

The solution can be written as

$$h(z) = \frac{\phi(z)}{z}\exp\left(-\frac{1}{z^\alpha}\right), \qquad (30)$$

where $\phi(z)$ obeys the simpler equation

$$\frac{d}{dz}\phi(z) = 2\alpha e^{1/z^\alpha}\int_0^z \frac{dx}{x^\alpha}h(x)h(z-x). \qquad (31)$$

An analytical solution of Eqs. (30) and (31) has not been found, but a numerical solution can be computed with the help of the following observations. First, it will appear that $\phi(z)$ is of order 1 in z for $z\sim 0$, and there is a factor e^{-1/z^α} in Eq. (30), so one can use the approximation

$$h(z) = 0 \quad \text{for } 0 \leq z \leq z_0, \qquad (32)$$

where z_0 depends on α. In practice $z_0 = (0.1)^{1/\alpha}$ appears to be a good choice since Eq. (32) is then fulfilled to within 10^{-4}. The effective reaction radius, which gives the average pair separation, is thus $z_0 t^{1/\alpha}$. With this approximation, the effective limits of the integral on the right-hand side of Eq. (31) are $z_0 \leq z \leq z - z_0$. Hence

$$\phi(z) = \phi(z_0), \quad h(z) = \frac{\phi(z_0)}{z}e^{-1/z^\alpha} \quad \text{for } z_0 \leq z \leq 2z_0. \qquad (33)$$

Integrating Eq. (31) we have

$$\phi(z) = \phi(z_0) + 2\alpha\int_{z_0}^z dx\, e^{1/x^\alpha}\int_{z_0}^{x-z_0}\frac{dy}{y^\alpha}h(y)h(x-y). \qquad (34)$$

Thus $\phi(z)$ in the range $2z_0 \leq z \leq 3z_0$ is determined by $h(z)$ in the range $z_0 \leq z \leq 2z_0$, as implied by Eq. (33). This procedure may be iterated to yield $h(z)$ and $\phi(z)$ for any z, once $\phi(z_0)$ is known.

In order to find $\phi(z_0)$, we observe that the large-z behavior of $\phi(z)$, given by Eq. (31), is $\phi(z) \sim z^\beta$, where β is the constant defined by

$$2\alpha\int_0^\infty h(z)\frac{dz}{z^\alpha} = \beta. \qquad (35)$$

On physical grounds, we expect that $\beta = 1$ in such a way that $h(z)$ reaches a finite value $h(\infty)$. This is indeed implied by constraint (15), which reads

$$w(r)\int_0^t \gamma(r,t')dt' = \omega^r \sim \frac{1}{2r} \quad \text{for } r < z_0 t^{1/\alpha}, \qquad (36)$$

where we have used relation (32) as in Sec. III. On writing Eq. (36) in the long-time limit as

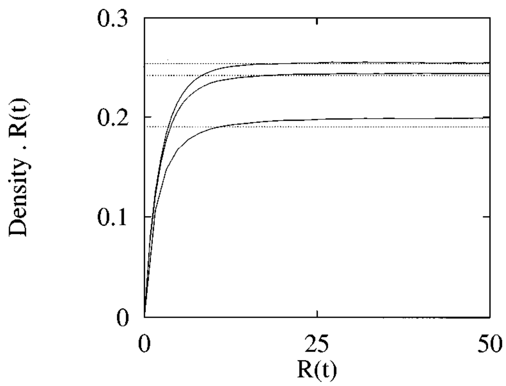

FIG. 2. Convergence of the density of surviving particles towards the asymptotic form (11): The product $R(t)\rho(t)$ in Monte Carlo simulations is compared with the asymptotes C_α (see the text) for $\alpha=6$, 4, and 2 from top to bottom.

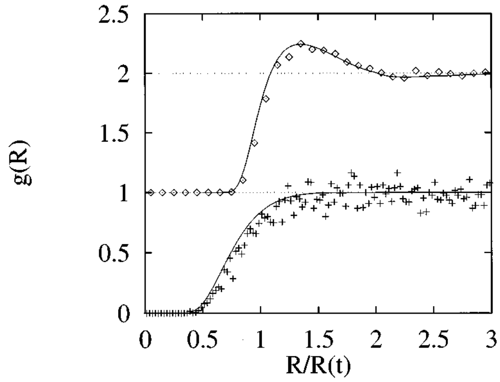

FIG. 3. Comparison of the asymptotic scaling form of the pair correlation from the Monte Carlo simulation and from the shielding approximation (smooth line) for $\alpha=6$ (top) and $\alpha=2$ (bottom).

$$2rw(r)\int_0^\infty \gamma(r,t)dt=1,$$

then using Eq. (28), and changing the variable from t to $z=rt^{-1/\alpha}$ at fixed r, this expression reads $\beta=1$. It is this constraint that determines $\phi(z_0)$. This is done numerically, and we further know that as α tends to ∞, we must have $\phi(z_0)=1/2$ in order for Eqs. (33) and (19) to coincide.

In practice, we proceed as follows for given α and z_0. We choose a trial value of $\phi(z_0)\sim 1/2$, construct $\phi(z)$ by iteration up to some $z=Nz_0$ in the asymptotic regime (N ranges from 8 to 10 for α ranging from 6 to 2), and adjust $\phi(z_0)$ until $\beta\sim 1$ [$\phi(z_0)$ appears to vary weakly, from 0.5 to 0.44 as α varies from 6 to 2]. Finally, we check the stability of the resulting $h(z)$ as z_0 is reduced and N is increased at constant $Nz_0=z$.

We have investigated the physically interesting integer values of α, $2\leqslant\alpha\leqslant 6$, with the results $h(\infty)=0.195$, 0.222, 0.242, 0.249, and 0.254. We recall from Eq. (11) that $\rho(t)\sim h(\infty)/t^{1/\alpha}$. Figure 2 illustrates this by comparing the product $\rho(t)t^{1/\alpha}$ from simulations with these predicted asymptotes. Figure 3 shows satisfactory agreement between the simulated and calculated correlation functions (in scaled units) for $\alpha=6$. The agreement for $\alpha=2$ is less satisfactory, as is the convergence of the analytical and simulated values of the product $\rho(t)t^{1/\alpha}$ in Fig. 2 because of numerical difficulties in both the estimation and the simulations. However, we note that the reaction front and the peak following it, indicating local ordering of surviving particles in the reactions with exponential [3] or $1/r^6$ coupling, are, as expected, less well defined or absent in the case $\alpha=2$.

V. CONCLUSION

Through Monte Carlo experiments we show that the shielding approximation is accurately fulfilled by the annihilation process. This approximation then closes the hierarchical equations, as in many other cases, like the trapping reaction $A+T\rightarrow T$ with immobile reactants and traps [8] or ballistic annihilation in a one-dimensional fluid [9]. In our case, the long-time solution of the resulting evolution equations has a scaling form involving the reaction radius in the expected way [10] and, as is well known, does not follow the mean-field regime. To the contrary, the evolution of the surviving population toward an ordered sate is particualrly striking for the exponential interaction, for which the correlation function can be predicted analytically. The local ordering is noticeable for multipolar interactions, especially $\alpha=6$, but decreases with α, practically disappearing for $\alpha=2$.

In conclusion, we would mention that this approach can be applied to the $A+B\rightarrow 0$ reaction, where it justifies the results obtained earlier with a staggered interaction model [11]. In higher dimensions, the shielding approximation may be replaced by a Kirkwood superposition approximation. As shown in [12], this leads to a closed system that can be solved numerically and appears to be in good agreement with the data at long times. The introduction of our techniques, especially the scaling forms, might lead to a more accurate or even an analytical solution for the exponential case.

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