

## Nonequilibrium density profiles of deposition-evaporation stochastic systems

M. D. Grynberg and R. B. Stinchcombe

*Theoretical Physics, Department of Physics, University of Oxford, 1 Keble Road, Oxford OX1 3NP, United Kingdom*

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We investigate the nonequilibrium kinetics of deposition and evaporation of dimers in bipartite lattices. For equal deposition-evaporation rates a quantum spin analogy enables one to describe the dimer stochastic evolution in terms of the isotropic spin- $\frac{1}{2}$  Heisenberg ferromagnet. This allows for an exact solution of nonequilibrium density profiles in higher dimensions starting from initially unstable states. It is shown that these systems exhibit diffusive behavior and how the interplay between jamming and dimensionality determines the type of asymptotic equilibration kinetics.

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There is much current interest in the theoretical understanding of stochastic cooperative systems evolving microscopically under conservative dynamical rules [1]. These systems are a common starting point to study a variety of physical situations with different relaxational processes and can provide valuable insight into nonequilibrium behavior and complex dynamics. Very recently, we have introduced a class of such models which describe fundamental aspects of deposition-evaporation (DE) processes [2]. The basic kinetic steps involve deposition without overlap and evaporation, possibly after reconstruction, of  $k$ -mers, where  $k = 1, 2, 3, \dots$  denotes monomers, dimers, trimers, etc. The underlying dynamics is directly related to lattice models of chemical reactions, ranging from catalysis processes to reactions on polymer chains [3].

The object of this work is to elucidate nonequilibrium dynamical aspects of the dimer model with equal DE rates. Our study is limited to the dimer system for two reasons. Firstly, already the case  $k = 2$  in bipartite lattices exhibits a Goldstone symmetry breaking largely responsible for the slow kinetics observed in more general situations. In particular, for equal DE rates the model is amenable to an exact calculation of nonequilibrium density profiles starting from initially unstable states. Secondly, for  $k > 2$  an infinite number of conservation laws partition the phase space into many universality classes thus posing rather prohibitive complex difficulties in the analysis of nonequilibrium dynamics [4].

It turns out that the stochastic time evolution generator of the dimer model can be cast in terms of an isotropic spin- $\frac{1}{2}$  Heisenberg ferromagnet whose global rotational symmetries we are going to exploit. The correspondence between these two types of problem enables us to explore the interplay between jamming (i.e., the inability to deposit or evaporate owing to the absence of two neighboring vacant or occupied sites), dimensionality, and their consequences in the stochastic nonequilibrium behavior. Although we shall consider hypercubic lattices throughout this work, generalizations to other bipartite geometries and to anisotropic rates should be obvious from the construction.

The microscopic dynamical rules of our dimer DE model are defined as follows. Pairs of nearest-neighbor sites of a  $d$ -dimensional lattice are selected at random

from  $N$  locations. A dimer deposition with rate  $\epsilon$  takes place if the chosen sites are both vacant. Alternatively, an evaporation attempt with rate  $\epsilon$  is successful provided the two selected sites are already occupied. If neither deposition nor evaporation is possible the sites' occupation numbers remain unchanged. Although this process does not include explicit particle hopping, note that an effective next-nearest-neighbor hopping, in which particle identity changes, can occur under two elementary DE steps. An example is dimer deposition onto two vacant sites (e.g.,  $j + 1, j + 2$ ) adjacent to a particle (at  $j$ ) followed by evaporation of the dimer now present on  $j, j + 1$ . Therefore our microscopic rules allow for reconstruction of dimers and redistribution of particles.

Starting from the master equation and representing a particle (vacancy) at site  $\mathbf{r}$  by the state of a pseudo-spin-operator  $\sigma_{\mathbf{r}}^z = +1$  (or  $-1$ ), the stochastic evolution of this system at time  $t$  is described by the action of the "Hamiltonian"  $\exp(-Ht)$ , namely,

$$H = -\epsilon \sum_{\mathbf{r}, j} [\sigma_{\mathbf{r}}^+ \sigma_{\mathbf{r}+\delta_j}^+ + \sigma_{\mathbf{r}}^- \sigma_{\mathbf{r}+\delta_j}^- - \frac{1}{2} (1 + \sigma_{\mathbf{r}}^z \sigma_{\mathbf{r}+\delta_j}^z)], \quad (1)$$

where  $\sigma_{\mathbf{r}}^+$  ( $\sigma_{\mathbf{r}}^-$ ) is a spin- $\frac{1}{2}$  raising (lowering) operator at site  $\mathbf{r}$  and  $j$  runs over all nearest-neighbor sites  $\mathbf{r} + \delta_j$  of  $\mathbf{r}$ . Deposition (evaporation) of dimers at rate  $\epsilon$  is described by the action of the first (second) term, whereas conservation of probability requires the appearance of a third (diagonal) term.

For *bipartite* lattices  $\Lambda = \Lambda_a + \Lambda_b$ , i.e., lattices with nonfrustrated Néel ordering, clearly the dynamics is partitioned into  $N + 1$  disconnected invariant subspaces as the DE process conserves the lattice staggered "magnetization"  $\sum_{\mathbf{r} \in \Lambda_a} \sigma_{\mathbf{r}}^z - \sum_{\mathbf{r} \in \Lambda_b} \sigma_{\mathbf{r}}^z$ . Therefore it is natural to introduce the sublattice mapping  $\tau_{\mathbf{r}} \equiv \sigma_{\mathbf{r}}$ ,  $\mathbf{r} \in \Lambda_a$ ,  $\tau_{\mathbf{r}} \equiv (\sigma_{\mathbf{r}}^x, -\sigma_{\mathbf{r}}^y, -\sigma_{\mathbf{r}}^z)$ ,  $\mathbf{r} \in \Lambda_b$  which transforms the evolution operator to a Heisenberg ferromagnet

$$\mathcal{H} = -\frac{\epsilon}{2} \sum_{\mathbf{r}, j} (\tau_{\mathbf{r}} \cdot \tau_{\mathbf{r}+\delta_j} - 1), \quad (2)$$

and leaves invariant all the components of the total angular momentum  $\mathbf{T} = \sum_{\mathbf{r}} \tau_{\mathbf{r}}$ . For unequal DE rates, in addition to an isotropic Heisenberg coupling,  $\mathcal{H}$  contains

staggered field and Dzyaloshinskii-Moriya terms [5], thus making the analysis rather involved. However, it is worth pointing out that within the *equilibrium* regime it is possible to obtain the asymptotic behavior of autocorrelation functions and the diffusion constants [2].

In the sublattice representation the steady state  $|\psi_m\rangle$  corresponding to the  $m$ -down spin sector can be obtained by rotating the fully jammed all spins up state  $|\psi_0\rangle$  (Néel configuration of the original system) by means of the lowering operator  $T^- = \sum_{\mathbf{r}} \tau_{\mathbf{r}}^-$ . Since  $[\mathcal{H}, T^-] = 0$ , a normalized steady state of this sector can be constructed as

$$|\psi_m\rangle = \frac{1}{m! \sqrt{\Omega}} (T^-)^m |\psi_0\rangle = \frac{1}{\sqrt{\Omega}} \sum_C |C\rangle, \quad (3)$$

where the sum runs over all the possible  $\Omega = \binom{N}{m}$  configurations  $|C\rangle = \tau_{\mathbf{r}_1}^- \cdots \tau_{\mathbf{r}_m}^- |\psi_0\rangle$  of the subspace within which the evolution takes place. The nature of the steady state has important consequences in the calculation of nonequilibrium properties. We first note that the probability of finding the system in a configuration  $|C\rangle$  at time  $t$  starting from an arbitrary initial configuration  $|C_0\rangle$  at  $t = 0$  is  $P_0(C, t) = \langle C | e^{-\mathcal{H}t} | C_0 \rangle$ , then it is clear from Eq. (3) that the nonequilibrium density profile  $\rho_0(\mathbf{r}, t)$  is given by

$$\rho_0(\mathbf{r}, t) = \sqrt{\Omega} \langle \psi_m | \hat{n}_{\mathbf{r}} e^{-\mathcal{H}t} | C_0 \rangle, \quad (4)$$

where  $\hat{n}_{\mathbf{r}} \equiv \tau_{\mathbf{r}}^+ \tau_{\mathbf{r}}^- = (1 + \tau_{\mathbf{r}}^z)/2$  is the occupation number operator at site  $\mathbf{r}$ . In the sublattice representation  $\hat{n}_{\mathbf{r}} \rightarrow (1 - \hat{n}_{\mathbf{r}})$  if  $\mathbf{r} \in \Lambda_b$ , although without loss of generality we may assume that  $\mathbf{r} \in \Lambda_a$ . Inserting a basis of common eigenstates  $\{|\mathbf{k}\rangle\}$  of  $\mathcal{H}$  and  $\mathbf{T}^2$  with eigenvalues  $\omega_{\mathbf{k}}, T(T+1)$ , respectively, it follows that

$$\rho_0(\mathbf{r}, t) = \sqrt{\Omega} \sum_{\mathbf{k}} e^{-\omega_{\mathbf{k}} t} \langle \psi_m | n_{\mathbf{r}} | \mathbf{k} \rangle \langle \mathbf{k} | C_0 \rangle. \quad (5)$$

However, from Eq. (3), the total spin of the steady state  $|\psi_m\rangle$  is  $T = N/2$ . Since the number operators  $\hat{n}_{\mathbf{r}}$  transform as tensors of rank 1, the Wigner-Eckart theorem ensures nonvanishing matrix elements  $\langle \psi_m | \hat{n}_{\mathbf{r}} | \mathbf{k} \rangle$  only if the total spin  $T$  of  $|\mathbf{k}\rangle$  and  $|\psi_m\rangle$  differs by 0 or 1, i.e.,  $T = N/2, N/2 - 1$ ; in either case with  $T^z = N/2 - m$ . Therefore the sum on the right-hand side of Eq. (5) is restricted to states  $|\mathbf{k}\rangle$  having  $T = N/2, N/2 - 1$ , a significant simplification. The contributing states are essentially rotated versions of single spin wave states  $|\varphi_{\mathbf{k}}\rangle = \sum_{\mathbf{r}} \varphi_{\mathbf{k}}(\mathbf{r}) \tau_{\mathbf{r}}^- |\psi_0\rangle$  [where  $\varphi_{\mathbf{k}}(\mathbf{r}) = N^{-1/2} e^{i\mathbf{k}\cdot\mathbf{r}}$  for a hypercubic lattice with periodic boundary conditions] and are the Goldstone modes of our dimer DE system. Recalling that the angular momentum algebra

imposes

$$T^- |T, T^z\rangle = \sqrt{(T+T^z)(T-T^z+1)} |T, T^z-1\rangle,$$

the  $|\mathbf{k}\rangle$  states are generated by applying  $m-1$  times ( $1 < m < N-1$ ) the lowering operator  $T^-$  to the spin waves  $|\varphi_{\mathbf{k}}\rangle$ . For  $\mathbf{k} \neq \mathbf{0}$  ( $T = N/2 - 1$ ) this yields

$$|\mathbf{k}\rangle = A_m (T^-)^{m-1} |\varphi_{\mathbf{k}}\rangle, \quad A_m = \sqrt{\frac{(N-m-1)!}{(N-2)!(m-1)!}}. \quad (6)$$

As was stated above, the state  $|\mathbf{k} = \mathbf{0}\rangle = |\psi_m\rangle$  has  $T = N/2$  and  $\omega_{\mathbf{0}} = 0$ , therefore from Eqs. (3) and (5) it can be seen that its contribution to  $\rho_0(\mathbf{r}, t)$  is

$$\sqrt{\Omega} \langle \psi_m | \hat{n}_{\mathbf{r}} | \psi_m \rangle \langle \psi_m | C_0 \rangle = \frac{1}{\Omega} \binom{N-1}{m} = 1 - m/N. \quad (7)$$

We are now left with the calculation of  $\langle \psi_m | \hat{n}_{\mathbf{r}} | \mathbf{k} \rangle$ ,  $\mathbf{k} \neq \mathbf{0}$ . This matrix element is expanded as

$$\langle \psi_m | \hat{n}_{\mathbf{r}} | \mathbf{k} \rangle = A_m \sum_{\mathbf{p}} \varphi_{\mathbf{k}}(\mathbf{p}) \langle \psi_m | \hat{n}_{\mathbf{r}} (T^-)^{m-1} \tau_{\mathbf{p}}^- |\psi_0\rangle. \quad (8)$$

On the other hand,

$$\begin{aligned} \hat{n}_{\mathbf{r}} (T^-)^{m-1} \tau_{\mathbf{p}}^- |\psi_0\rangle &= (m-1)! (1 - \delta_{\mathbf{r}, \mathbf{p}}) \\ &\times \sum_{j_1 < \dots < j_{m-1}}'' \tau_{\mathbf{r}_{j_1}}^- \cdots \tau_{\mathbf{r}_{j_{m-1}}}^- \tau_{\mathbf{p}}^- |\psi_0\rangle, \end{aligned} \quad (9)$$

where the double prime restricts the sums to vectors  $\mathbf{r}_j \neq \mathbf{r}, \mathbf{p}$ . For  $\mathbf{r} \neq \mathbf{p}$  there are  $\binom{N-2}{m-1}$  terms contributing equally to  $\langle \psi_m | \hat{n}_{\mathbf{r}} | \mathbf{k} \rangle$ . Hence we obtain

$$\begin{aligned} \langle \psi_m | \hat{n}_{\mathbf{r}} | \mathbf{k} \rangle &= (m-1)! \binom{N-2}{m-1} \frac{A_m}{\sqrt{\Omega}} \\ &\times \sum_{\mathbf{p}} \varphi_{\mathbf{k}}(\mathbf{p}) (1 - \delta_{\mathbf{r}, \mathbf{p}}) \\ &= -\sqrt{\frac{m(N-m)}{N(N-1)}} \varphi_{\mathbf{k}}(\mathbf{r}). \end{aligned} \quad (10)$$

To complete the calculation of nonequilibrium density profiles we now evaluate  $\langle \mathbf{k} | C_0 \rangle$ . First we note that for  $\mathbf{k} \neq \mathbf{0}$

$$|\mathbf{k}\rangle = (m-1)! A_m \sum_{\mathbf{r}} \varphi_{\mathbf{k}}(\mathbf{r}) \sum_{j_1 < \dots < j_{m-1}}' \tau_{\mathbf{r}_{j_1}}^- \cdots \tau_{\mathbf{r}_{j_{m-1}}}^- \tau_{\mathbf{r}}^- |\psi_0\rangle, \quad (11)$$

where the prime restricts the sums to vectors  $\mathbf{r}_j \neq \mathbf{r}$ . For a given set  $\{\mathbf{r}_1^0, \dots, \mathbf{r}_m^0\}$  of initial coordinates of  $\tau$  particles in  $|C_0\rangle = \tau_{\mathbf{r}_1^0}^- \cdots \tau_{\mathbf{r}_m^0}^- |\psi_0\rangle$ , it follows from Eq. (11) that

$$\langle \mathbf{k} | C_0 \rangle = (m-1)! A_m \sum_{i=1}^m \varphi_{\mathbf{k}}^*(\mathbf{r}_i^0), \quad \mathbf{k} \neq \mathbf{0}. \quad (12)$$

Therefore, recalling Eqs. (7), (10), and (12), the density

profile turns out to be

$$\rho_0(\mathbf{r}, t) = 1 - \sum_{i=1}^m \sum_{\text{all } \mathbf{k}} e^{-\omega_{\mathbf{k}} t} \varphi_{\mathbf{k}}(\mathbf{r}) \varphi_{\mathbf{k}}^*(\mathbf{r}_i^0), \quad \mathbf{r} \in \Lambda_a. \quad (13)$$

For a  $d$ -dimensional hypercubic lattice with  $N = L_1, \dots, L_d$  sites and periodic boundary conditions, the Goldstone excitations are of the form  $\omega_{\mathbf{k}} = 2\epsilon \sum_{j=1}^d (1 - \cos k_j)$  with  $k_j = 2\pi n/L_j$  ( $0 \leq n < L_j$ ), hence in the limit  $L_j \rightarrow \infty$  we finally obtain

$$\rho_0(\mathbf{r}, t) = 1 - \exp(-2d\epsilon t) \sum_{i=1}^m \prod_{j=1}^d I_{n_j - n_{i,j}}^0(2\epsilon t), \quad \mathbf{r} \in \Lambda_a \quad (14)$$

where  $I_n(z)$  is a modified Bessel function of integer order [6], whereas  $n_j - n_{i,j}^0$  denotes the distance of site  $\mathbf{r}$  to  $\mathbf{r}_i^0$  measured along the  $j$  direction (the lattice constant has been taken as unity). For  $\mathbf{r} \in \Lambda_b$  the above expression should be modified as  $1 - \rho_0(\mathbf{r}, t)$ . Consequently, it follows that *within* the sublattice representation the dimer DE model exhibits a *diffusive* dynamics as Eq. (14) is the discrete version solution of a  $d$ -dimensional diffusion equation  $\partial \tilde{\rho}(\mathbf{r}, t)/\partial t = \epsilon \nabla^2 \tilde{\rho}(\mathbf{r}, t)$  with  $\tilde{\rho}(\mathbf{r}, t) = 1 - \rho(\mathbf{r}, t)$  for  $\mathbf{r} \in \Lambda_b$ .

We now consider the implications of our results for the nonequilibrium dimer kinetics arising from either non-jammed or partially jammed initial conditions. A quantitative measure of jamming in the initial state is given by  $J_0 \equiv 1 - \langle C_0 | \mathcal{H} | C_0 \rangle / N_b$  as the diagonal elements of  $\mathcal{H}$  count the total number of ways in which a given state can evolve to different configurations in a single elementary DE process (at most  $N_b$ , the number of lattice bonds). For instance, an initially empty substrate (Néel state in the sublattice spin representation) has minimum jamming  $J_0 = 0$ . This is a very common situation within the context of cooperative sequential adsorption [7], thus it is of interest to elucidate the resulting dynamics of equilibration. It can be easily checked from Eq. (14) that  $\rho_0(\mathbf{r}, t)$  is, as expected, translationally invariant and relaxes *exponentially* in time as  $\rho(t) = (1 - e^{-t/\tau_d})/2$  with a characteristic relaxation time  $\tau_1 = (4\epsilon)^{-1}$  for  $d = 1$ , and  $\tau_2 = (8\epsilon)^{-1}$  in the two-dimensional case.

In contrast, the role of dimensionality is crucial for almost unjammed initial conditions. Consider, for example, the case of an initial shock profile  $\rho(n, 0) = 1$  for  $n \leq 0$  and  $\rho(n, 0) = 0$  for  $n > 0$ , where  $n$  denotes the distance to the shock front or interface. In the sublattice spin language this corresponds to a straight domain wall dividing two antiferromagnetic domains of opposite orientation. Here the jamming is almost minimum ( $J_0 = 0^+$ ), however sufficient to yield substantial differences. On different grounds, the case of initial shock fronts is particularly important to analyze within our microscopic approach as in this situation the gradients of macroscopic variables (such as density) diverge and the hydrodynamic equations break down [8].

The dimer character of our DE model imposes a distinction between even and odd  $n$  distances although in either case the asymptotic kinetics is the same, so for simplicity we may consider even distances, say. For the one-dimensional lattice Eq. (14) yields

$$\rho(2n, t) = \rho(0, t) + e^{-2\epsilon t} F_n(t), \quad n \neq 0 \quad (15)$$

where  $\rho(0, t) = [1 + e^{-2\epsilon t} I_0(2\epsilon t)]/2$  and  $F_n(t)$  is defined as

$$F_n(t) = \sum_{j=1}^{-n} [I_{2j}(2\epsilon t) - I_{2j-1}(2\epsilon t)], \quad n < 0 \\ = - [F_{-n}(t) + I_0(2\epsilon t) - I_{2n}(2\epsilon t)], \quad n > 0. \quad (16)$$

In the limit  $n \rightarrow \pm\infty$ ,  $t \rightarrow \infty$  with  $n^2/t$  held finite, we obtain

$$\rho(2n, t) \simeq \frac{1}{2} + \frac{1}{4\sqrt{\pi\epsilon t}} \exp\left(-\frac{n^2}{\epsilon t}\right). \quad (17)$$

Therefore the system is dominated by a *slow* relaxation dynamics  $\sim t^{-1/2}$  and exhibits a diffusive interface whose width grows as  $4\sqrt{\epsilon t}$ . In contrast, the situation in the two-dimensional case changes dramatically. From Eq. (14) it is straightforward to show that

$$\rho(2n, t) = \frac{1}{2} + e^{-6\epsilon t} \left[ \frac{1}{2} I_0(2\epsilon t) + F_n(t) \right], \quad |n| > 2. \quad (18)$$

Thus, in the long-time limit we find that the asymptotic kinetics towards equilibrium is characterized by a *fast* (exponential) decay, namely

$$\rho(2n, t) \simeq \frac{1}{2} + \frac{1}{4\sqrt{\pi\epsilon t}} \exp\left[-\left(4\epsilon t + \frac{n^2}{\epsilon t}\right)\right]. \quad (19)$$

The reason for this rather different behavior is that in  $d = 2$  the direction parallel to the interface is initially unjammed. That is the active region which ultimately gives rise to the fast dynamics obtained above.

Jammed environments play a central role in the origin of slow equilibration kinetics, particularly in higher dimensions. To illustrate this point we finally consider the complementary scenario in which the only initially active region is the boundary between two antiferromagnetic domains of opposite orientation (i. e., a shock profile in the sublattice spin representation). In contrast with the situations analyzed so far the amount of initial jamming is almost maximum ( $J_0 = 1^-$ ). In Fig. 1(a) we show a snapshot of the dimer DE stochastic evolution at  $\epsilon t = 20$ . For displaying purposes we only plot the particles on one sublattice. The nonequilibrium density profile and asymptotic behavior can be straightforwardly worked out from Eq. (14). The result turns out to be

$$\rho(n, t) = \rho(0, t) + e^{-2\epsilon t} \sum_{j=1}^{-n} I_j(2\epsilon t), \quad n < 0 \\ = 1 + e^{-2\epsilon t} I_n(2\epsilon t) - \rho(-n, t), \quad n > 0, \quad (20)$$

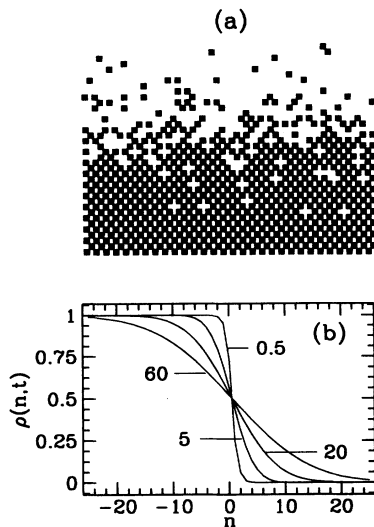


FIG. 1. (a) Snapshot of dimer deposition-evaporation stochastic evolution at  $\epsilon t = 20$  starting from the nearly jammed background referred to in the text (only particles in one sublattice are shown) and (b) corresponding nonequilibrium density profiles.

where  $\rho(0, t)$  is taken as in Eq. (15), whereas  $n$  denotes the distance to the initial unjammed interface for sites belonging to one sublattice [see Fig. 1(b)]. Remarkably, this result is *independent* of the spatial dimensionality. In the long-time limit with  $n^2/t$  kept finite the profile can be written as

$$\rho(n, t) \simeq \frac{1}{2} \left[ 1 - \operatorname{erf} \left( \frac{n}{\sqrt{4\epsilon t}} \right) \right], \quad (21)$$

where  $\operatorname{erf}(z) = (2/\sqrt{\pi}) \int_0^z e^{-u^2} du$ . Since  $\operatorname{erf}(z) \simeq 2z/\sqrt{\pi}$  for  $z \ll 1$  [6], the dimer system is dominated by a *slow* asymptotic kinetics  $\sim t^{-1/2}$  and develops a partially jammed interface of width  $2\sqrt{\pi\epsilon t}$ . This indicates that in higher dimensions at least partially jammed initial conditions along *all* principal  $d$  directions can lead to a slow equilibration dynamics.

In conclusion, we have introduced a spin operational formalism which enables one to elucidate nonequilibrium aspects of DE stochastic systems recently introduced in the literature. For equal DE rates, conservation of probability imposes a full rotational symmetry in the evolution operator which ultimately allows for an exact solution in higher dimensions. This rather crucial feature is lost for unequal DE rates. It should be emphasized that the above results do not constitute a complete solution of the dimer DE model. Information about nonequilibrium spatial pair correlations is also of interest. Such correlations could be calculated after shifting the problem (by spin rotation) to the two *interacting* magnon sector. The possibility of bound states arising in this sector could contribute importantly to the long-time kinetics of these correlations and therefore to the structure factors. Studies in that direction are under consideration. The elucidation of nonequilibrium dynamics for the general  $k$ -mer system remains quite open.

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