Annihilating random walks in one-dimensional disordered media

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We study diffusion-limited pair annihilation $A+A\to 0$ on one-dimensional lattices with inhomogeneous nearest-neighbor hopping in the limit of an infinite reaction rate. We obtain a simple exact expression for the particle concentration $\rho_k(t)$ of the many-particle system in terms of the conditional probabilities $P(m;t|l;0)$ for a single random walker in a *dual* medium. For some disordered systems with an initially randomly filled lattice this leads asymptotically to $\overline{\rho(t)}=\overline{P(0;2t|0;0)}$ for the disorder-averaged particle density. We also obtain interesting exact relations for single-particle conditional probabilities in random media related by duality, such as random-barrier and random-trap systems. For some specific random-barrier systems the Smoluchovsky approach to diffusion-limited annihilation turns out to fail. [S1063-651X(98)13702-9]

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Stochastic reaction-diffusion processes play an important role in the description of interacting many-particle systems in both physics and chemistry. Usually real systems are much too complex to be amenable to analytical or even numerical investigation. However, particularly in the context of critical phenomena, simple toy models may suffice to determine universal properties correctly and to predict and explain observed power laws or universal amplitude ratios. Hence it is of importance both to examine the behavior of such models and to understand possible relationships between microscopically different processes and their characterization in terms of universality classes. In this paper we investigate a model of annihilating random walkers describing diffusion-limited pair annihilation (DLPA) of identical particles in inhomogeneous media. This process describes both chemical reactions where the particles change their state into an inert reaction product which takes no part in the subsequent dynamics of the system, or physical reactions where the particles actually annihilate under the emission of radiation. For a recent review of experimental and theoretical applications of the ordered model, see Refs. [1,2]. The model is related by a similarity transformation to the diffusion-limited coagulation process [3,4] which describes, e.g., laser-induced exciton dynamics on polymers. The model also maps to Glauber dynamics for the one-dimensional Ising model [5–7], and is therefore of interest in the study of the kinetics of disordered equilibrium systems. It plays an important role not only for the study of spin-relaxation phenomena but also for the solution of more intricate problems such as the derivation of persistence exponents [8] and nonequilibrium steady states [9,10].

The first comprehensive treatment of homogeneous diffusion-limited annihilation dates back to Smoluchovsky's classical work in 1917 [11], but, despite renewed strong interest in the 1980s and 1990s the experimentally more realistic case of spatially inhomogeneous particle hopping rates, e.g., in the presence of quenched disorder, has so far received very little attention [4,12,13]. In homogeneous, translationally invariant environments the particle density decays with a power law which depends on the dimensionality of the system. Both theoretically and experimentally one finds $\rho(t)\sim 1/\sqrt{Dt}$ in one dimension, in agreement with exact results [14,15]. Interestingly, this result is at variance with the (dimensionality-independent) mean-field behavior $\rho(t)\sim 1/(Dt)$ which is correct only in three (and higher) dimensions. The amplitude is universal in the sense that it depends neither on the initial density (for random initial conditions) nor on the reaction rate [16]. However, if particles are moving in a nontranslationally invariant energy landscape, it is not obvious how this will change the decay of the local or overall particle concentration. The physical motivation behind the study of the one-dimensional case is not only its experimental relevance for polymer physics, but also its theoretical importance in the understanding of the role of fluctuations in low-dimensional systems. In both one- and two-dimensional systems, diffusive mixing is inefficient and leads to the building up of large-scale correlations. Thus the classical mean-field rate equations for the study of these systems tend to fail, and require a more sophisticated treatment.

Before going into detail, we remind the reader of the essentials of Smoluchovsky's reasoning. The idea behind this approach [11] is to replace in the mean-field rate equation for the density $\dot{\rho}(t)=-\lambda\rho^2(t)$, the reaction constant λ by an effective time-dependent reaction rate which is proportional to the diffusive current $j(t)$ into an absorbing particle in a background of constant density. In one dimension, where $j(t)\propto\sqrt{D/t}$, this leads (up to the universal amplitude which cannot be determined from the Smoluchovski argument) to $\rho(t)\propto 1/\sqrt{Dt}$. One would like to know whether the fluctuation-improved mean-field theory of Smoluchovsky which predicts the correct behavior of the one-dimensional ordered system remains valid in the presence of disorder. Naively, one might expect that the diffusion constant of the pure system would have to be replaced by some effective diffusion constant of the disordered system, i.e., $\overline{\rho(t)}\propto 1/\sqrt{D_{\rm eff}t}$ for the disorder-averaged density. But clearly this cannot always work, as can be seen in a simple and natural example: Suppose one investigates DLPA on an ensemble of ordered chains of varying length $L\leqslant\Lambda$, modeling, e.g., a polymer mixture of polymer fragments of varying finite length. This is equivalent to taking an infinite chain and place randomly, but with maximal distance Λ, broken bonds across which particles cannot move. If initially particles are

placed randomly with probability 1/2 on each lattice site, then the steady state density in each chain of length L is equal to $\rho^*=1/(2L)$. Assuming that each chain length occurs with equal probability, then the averaged density $\overline{\rho^*}=\overline{1/(2L)}\sim\ln\Lambda/(2\Lambda)$ for large maximal length Λ. On the other hand, since the particle is confined to a box of length L the system is subdiffusive with $D_{\rm eff}\propto\Lambda^2/t$. Hence the Smoluchovsky formula gives the wrong result $\overline{\rho^*}\propto 1/\Lambda$. Even worse, with this Smoluchovsky approach one cannot even estimate the approach of the density to its stationary value. One could, of course, try to be smart and apply Smoluchovsky's approach to a finite system and then average over system size. This indeed gives $\overline{\rho^*}\propto\ln\Lambda/(\Lambda)$. However, if applied to a different system where one distributes infinitely deep traps (sites out of which particles cannot jump) at a maximal distance Λ, then this refined Smoluchovsky argument would also give $\overline{\rho^*}\propto\ln\Lambda/(\Lambda)$ for this model. However, the exact result given below shows that, in fact, for this type of disorder, $\overline{\rho^*}=1/\Lambda$, which happens to be consistent with the first, naive Smoluchovsky result. We conclude that there is no simple argument which tells one how to proceed without already knowing the answer. Thus exact results are required for a study of DLPA in inhomogeneous media.

To this end we investigate, by a new exact mapping, a DLPA lattice model with space-dependent hopping rates. The particles have no attractive or repulsive interaction between themselves; they hop with fixed rates r_k (ℓ_k) from lattice site k to site $k+1$ ($k-1$). When two particles meet on site k they both annihilate instantaneously. This limit of an infinite reaction rate corresponds to the renormalization group fixed point of the ordered system [2,16,17], and we believe that the disordered system with a finite reaction rate will also be in the same universality class as the infinite rate limit.

In the case of constant hopping rates the predictions of this model are in excellent agreement with experimental data on exciton dynamics on very long ordered polymer chains. Thus we expect that the disordered model gives an equally good description of the behavior of realistic, disordered systems.

We define the process in terms of a master equation for the probability $P(\eta;t)$ of finding, at time t, a configuration η of particles on a lattice of L sites. Using standard techniques [18–20], we express the time evolution given by the master equation in terms of a quantum Hamiltonian H. Since particles annihilate instantaneously when they meet, there can never be more than one particle on any given site. The idea is now to extend the bosonic Fock space formalism of [18–20] to a quantum spin chains representation [21–23]. One represents each of the 2^L possible particle configurations η by a vector $|\eta\rangle$, which, together with the transposed vectors $\langle\eta|$, form an orthonormal basis of a vector space $X=(\mathbb{C}^2)^{\otimes L}$. A state η with N particles placed on sites $k_1,\ldots,k_N$ is represented by $|k_1,\ldots,k_N\rangle$, the completely empty lattice by the vector $|0\rangle$. The probability distribution corresponds to a state vector $|P(t)\rangle=\Sigma_{\eta\in X}P(\eta;t)\,|\eta\rangle$, and one writes the master equation in the form

$$\frac{d}{dt}P(\eta;t)=-\langle\eta|H|P(t)\rangle\,, \tag{1}$$

where the off-diagonal matrix elements of H are the (negative) transition rates between states, and the diagonal entries are the inverse of the exponentially distributed lifetimes of the states. A distribution at time t is given in terms of an initial state at time $t=0$ by $|P(t)\rangle=e^{-Ht}\,|P(0)\rangle$. The expectation value $\rho_k(t)=\langle s|n_k\,|P(t)\rangle$ for the density at site k is given by the projection operator n_k which has value 1 if there is a particle at site k and 0 otherwise. The vector $\langle s|=\Sigma_{\eta\in X}\langle\eta|$ performs the average over all possible final states of the stochastic time evolution. Choosing the basis of X such that a particle (vacancy) on site k corresponds to spin up (down) the Hamiltonian

$$H=-\sum_k\,(r_k h_k^+ +\ell_k h_k^-) \tag{2}$$

for the process can be written in terms of Pauli matrices $h_k^\pm=(s_k^+ s_{k\pm1}^- + s_k^+ s_{k\pm1}^+ - n_k)$, where $n_k=(1-\sigma_k^z)/2$ and $s_k^\pm=(\sigma_k^x\pm i\sigma_k^y)/2$ create (s_k^-) and annihilate (s_k^+) particles (see Ref. [4] for details). Since the time evolution conserves particle number modulo 2, it is convenient to work only on the even and odd subspaces defined by the projector $P^\pm=(1\pm Q)/2$ where $Q=(-1)^N=\Pi_k\sigma_k^z$. For averaging over final states we then use $\langle s^\pm|=\langle s|P^\pm$. The projection on the even sector of the uncorrelated initial state with a density $\frac{1}{2}$ used below is given by the vector $|\tfrac{1}{2}^+\rangle=(\tfrac{1}{2})^{L-1}\,|s^+\rangle$.

In one dimension with *homogeneous* nearest neighbor hopping, DLPA is related to zero-temperature Glauber dynamics by a domain-wall duality transformation [6] which is an invertible similarity transformation [7]. On the other hand, zero-temperature Glauber dynamics can be brought by another similarity transformation into a form which is the *transpose* of the Hamiltonian for (homogeneous) DLPA [24]. We use these results to construct a new matrix $\mathcal{D}$ such that the process defined by $\hat{H}=\mathcal{D}^{-1}H^T\mathcal{D}$ also describes a DLPA process with nearest neighbor hopping, albeit with different hopping rates in the presence of disorder. For the even particle sector we find

$$\mathcal{D}_+=\gamma_1\gamma_2\cdots\gamma_{2L-1}\,, \tag{3}$$

where $\gamma_{2k-1}=[(1+i)\sigma_k^z-(1-i)]/2$, $\gamma_{2k}=[(1+i)\sigma_k^x\sigma_{k+1}^x-(1-i)]/2$, and $\mathcal{D}_-=-\mathcal{D}_+\sigma_L^x$ for the odd particle sector. To see what happens under the mapping, we note that $\mathcal{D}_\pm$ is unitary and transforms Pauli matrices as follows:

$$\mathcal{D}_\pm^{-1}\sigma_k^x\sigma_{k+1}^x\mathcal{D}_\pm=\begin{cases}\sigma_k^z & k\neq L\\ Q\sigma_L^z & k=L\,,\end{cases} \tag{4}$$

$$\mathcal{D}_\pm^{-1}\sigma_{k+1}^z\mathcal{D}_\pm=\begin{cases}\sigma_k^x\sigma_{k+1}^x & k\neq L\\ \pm Q\sigma_L^x\sigma_1^1 & k=L\,.\end{cases} \tag{5}$$

In the even sector one now finds

$$\hat{H}=-\sum_k\,(r_k h_k^- +\ell_k h_{k-1}^+). \tag{6}$$

This process is of the same form as the original process (2), but with dual hopping rates $\hat{\ell}_k = r_k$, $\hat{r}_k = \ell_{k+1}$. We shall refer to the environment defined by the dual rates as the dual environment [25].

In order to make use of the mapping, one needs to know how a given initial distribution and the observables change under the transformation. For the transformation laws for states, one needs Eqs. (4) and (5) and $\mathcal{D}_+^{-1}|s^+\rangle = -i(i-1)^{L-1}|0\rangle$, $\mathcal{D}_+|s^+\rangle = i(-i-1)^{L-1}|0\rangle$ for the even sector, and analogous relations for the odd sector. For the density at site k for an arbitrary initial state, one then finds

$$\langle s^+|n_k e^{-Ht}|P_0\rangle = \tfrac{1}{2}\langle P_0|\mathcal{D}e^{-\hat{H}t}(1-\sigma_{k-1}^x\sigma_k^x)\mathcal{D}^{-1}|s^+\rangle. \tag{7}$$

The transformed initial state is a superposition of the steady state (the empty lattice) and the two-particle state with particles at sites $k-1,k$. Bearing in mind that $\hat{H}$ does not have any particle creation terms one now realizes that the time dependence of the density for an arbitrary many-particle initial distribution is completely given by the dynamics of just *two* annihilating random walkers in the dual disordered environment, $\rho_k(t) = \alpha - \Sigma_{m,l} a_{ml}\langle m,l|e^{-\hat{H}t}|k-1,k\rangle$, where the coefficients α and a_{lm} are determined by the initial state and straightforward to work out [26].

In order to avoid immaterial technical complications with boundary terms, from now on we consider only infinite systems. By choosing some of the hopping rates equal to zero one can always recover results for finite systems. To calculate the two-particle transition probability $\hat{P}(m,n;t|k,l;0) = \langle m,n|e^{-\hat{H}t}|k,l\rangle$, we note that the transition probability for a single random walker $\hat{P}(m,k;t) \equiv \hat{P}(m;t|k,0)$ is the sum over all paths leading from k to m, each weighted with its proper statistical weight given by the hopping rates and the particular form of the trajectory. Hence for two noninteracting particles moving from k to m and from l to n, respectively, $\hat{P}(m,n;t|k,l;0) = \hat{P}(m,k;t)\hat{P}(n,l;t)$. This sum includes the contribution of paths which cross each other. In an annihilating random walk of otherwise noninteracting particles, the contribution of all crossing paths have to be subtracted. Since we are on a one-dimensional lattice this contribution is just the one given by all paths which start at site k and end at site n (instead of m), and which start at site l and end at site m (instead of n). Therefore,

$$\hat{P}(m,n;t|k,l;0) = \hat{P}(m,k;t)\hat{P}(n,l;t) - \hat{P}(n,k;t)\hat{P}(m,l;t). \tag{8}$$

This further reduces the calculation of the density to the solution of a *single-particle* random-walk problem in the dual random environment. For an uncorrelated random initial state with density $\frac{1}{2}$ in the sector of even particle number, one obtains

$$\rho_k(t) = \langle 2|e^{-\hat{H}t}|k-1,k\rangle/2, \tag{9}$$

where $\langle 2| = \Sigma_{n>m}\langle m,n|$ is the sum over all states with two particles. Thus the density at site k is equal to one half the survival probability of two annihilating random walkers starting at sites $k-1,k$ and moving in the dual environment. As a first specific result we calculate the final density of a system with infinitely deep traps placed randomly, as discussed above. Clearly, if k is not a trap site, then $\rho_k(\infty) = 0$. Hence the disorder-averaged density $\overline{\rho(\infty)} = q\widetilde{\rho}(\infty)$ where $q = 2/\Lambda$ is the density of traps and $\widetilde{\rho}(t)$ is the density at a trap site. From Eq. (9) one finds $\widetilde{\rho}(t) = \frac{1}{2}$, and therefore $\overline{\rho(\infty)} = 1/\Lambda$.

Consider now the relation between the single-particle conditional probabilities for dual environments. One may write $P(m;t|k;0) = \langle s|^{\text{odd}}\Pi_{i=1}^{m-1}\sigma_i^z n_m e^{-Ht}|k\rangle$. By taking the transpose and transforming under $\mathcal{D}_-$, one obtains in the infinite volume limit, the interesting exact relation

$$P(m,k;t) - P(m,k-1;t) = \hat{P}(m-1,k-1;t) - \hat{P}(m-1,k;t). \tag{10}$$

Note that Eqs. (7)–(10) hold for any fixed hopping environment, disordered or inhomogeneous, but regular. In this paper we focus on disordered systems with translationally and reflection invariant hopping rate distributions. In this case, Eq. (10) gives

$$\overline{P(r;t)} = \overline{\hat{P}(r;t)} + c(t), \tag{11}$$

with an undetermined function $c(t)$ which is irrelevant for what follows. Relations (10) and (11) are remarkable in that they relate the conditional probabilities for dual systems.

In order to analyze Eq. (9) further, we take a mean-field approach to the disorder average, i.e., we replace in Eq. (8) the disorder average of the conditional probability for two distinguishable noninteracting random walkers $\overline{\hat{P}(m,k;t)\hat{P}(n,l;t)}$ by the factorized average $\overline{\hat{P}(m,k;t)}\,\overline{\hat{P}(n,l;t)}$.

We are convinced numerically that this factorization holds well for the random-barrier model with uncorrelated bond hopping probabilities b_k drawn uniformly from the interval $0.05 < b_k \leqslant \frac{1}{2}$. This was done by an exact numerical solution of the discrete-time master equation for a random walker on a lattice of $L = 200$ sites for a given random realization of the disorder, and then taking the average over 100 000 disorder realizations (to keep disorder-related fluctuations small). To show this factorization in Fig. 1, the function

$$R_{nl}^{mk}(t) = \frac{\overline{P(m,k;t)\hat{P}(n,l;t)}}{\overline{\hat{P}(m,k;t)}\,\overline{\hat{P}(n,l;t)}} - 1 \tag{12}$$

is shown for $m = n = 1$ and $k = l = 0$ in a double logarithmic plot. The return probability gives the largest contribution to the function $R_{nl}^{mk}(t)$, hence we do not need to check the factorization at other positions. The function $R_{10}^{10}(t)$ is fitted well by a power law

$$R_{10}^{10}(t) \sim t^{-\alpha}, \tag{13}$$

where $\alpha = -0.49$. For this the fit routine of the software package MATHEMATICA was used. We expect that the exact

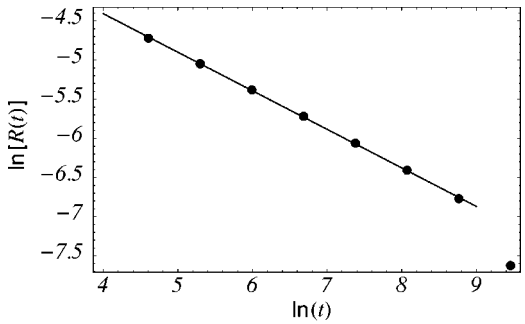

FIG. 1. Factorization of one-particle condition probabilities after the disorder average. $R(t)$ vs t is shown in a double-logarithmic plot. The line has a slope -0.49, and is a fit of the first seven points which are produced by exact numerical calculations of the master equation where the jump probabilities b_k were taken randomly from a uniform distribution between $0.05<b_k\leqslant\frac{1}{2}$.

exponent to be $\alpha=\frac{1}{2}$. The deviation of the last point in Fig. 1 from the line originates from finite-size effects. The square root of the time at this point is about the length of the system, and therefore the constant stationary probability of the random-barrier system is reached exponentially fast. We also investigated the case where zero is the lower limit of the distribution of the hopping probabilities in the same system as above. In this case the behavior of the mean square displacement of a single particle is subdiffusive $\langle x^2\rangle\sim t/\ln(t)$ [27,30]. This is due to the existence of very small hopping probabilities in even very small regions in the chain. The factorization can only hold for very long times. Nevertheless the function $R_{nl}^{mk}(t)$ is very small even for short times and for longer times there is a tendency of the function $R_{nl}^{mk}(t)$ to decrease as it can be seen in Fig. 2, where $R_{nl}^{mk}(t)$ is spread vs $\ln(t)$. Independent arguments for the validity of this assumption for more general types of disorder are given in the conclusions.

Then, with Eqs. (9) and (11), the disorder average of the density for the initially randomly filled lattice is given by

$$\overline{\rho(t)}=\overline{(P(0;2t)}+\overline{P(1;2t)}\,. \tag{14}$$

For large times, Eq. (14) becomes the return probability quoted in the Abstract. Having in mind processes like exciton dynamics it is reasonable to consider (A) random-bond

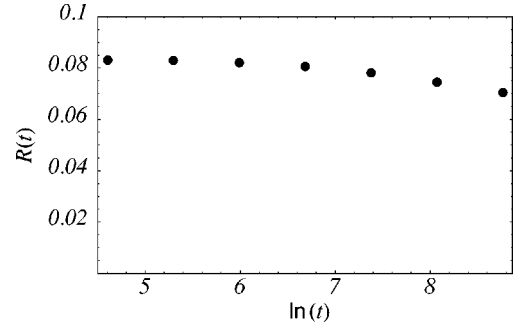

FIG. 2. Factorization of one-particle condition probabilities after the disorder average. $R(t)$ vs $\ln(t)$. The points are produced as in Fig 1, but the jump probabilities b_k were taken randomly from a uniform distribution between $0<b_k\leqslant\frac{1}{2}$.

disorder $r_k=\ell_{k+1}\equiv b_k$ where hopping across a bond $k,k+1$ is symmetric, but bond-dependent, and (B) random-site disorder $r_k=\ell_k\equiv s_k$. The energy of a particle in the random-barrier model (A) is the same at each site, but between sites there are energy barriers of random height E_k. Thermal fluctuations cause the particle to jump over these barriers with a random rate $b_k\propto\exp(-\beta E_k)$. Case (B) corresponds to the random trap model. Here the particle sits in a site-dependent potential of depth $-E_k$. Random-bond and random-site disorders are dual in the sense of Eq. (6). Since for the random-bond model $P(m,k;t)=P(k,m;t)$, we conclude that the disorder-averaged conditional probabilities of the random-barrier model and the random-trap model are equal in one dimension for any translationally invariant disorder distribution up to a function $c(t)$. For an uncorrelated ergodic disorder distribution, diffusion in random barrier systems converges to Brownian motion, i.e., the averaged conditional probability becomes asymptotically equal to a Gauss distribution with an effective diffusion constant [27–30]

$$D_{\rm eff}^{-1}=\overline{b_k^{-1}}, \tag{15}$$

and therefore, asymptotically,

$$\overline{\rho(t)}=\frac{1}{\sqrt{4\pi D_{\rm eff}t}} \tag{16}$$

for random-trap and random-barrier systems. For exponentially distributed barrier or trap energies $\nu(E)\propto\exp(-E/\sigma)$ the random walk becomes subdiffusive below a critical temperature given by $\sigma\beta_c=1$ [31,32]. This leads to a time-dependent effective diffusion coefficient $D_{\rm eff}\propto t^{(1-\sigma\beta)/(1+\sigma\beta)}$, and hence to a slower nonuniversal power law decay of the density $\rho(t)\propto t^{-1/(1+\sigma\beta)}$.

To conclude, we obtained the following results.

(i) We found the duality relations (10) and (11) for random walkers in dual systems. They play a role in the derivation of Eqs. (14)–(16), but are also of interest in their own right.

(ii) The Smoluchovsky approach to DLPA is consistent with random-trap and random-bond systems with vanishing broken bond probability [Eqs. (15) and (16)], but fails for some important disordered broken bond systems in one dimension. This raises the question under which general conditions on the disorder of the Smoluchovsky approach is correct (see below).

(iii) The expectation value of the density in diffusion-limited annihilation in one dimension with nearest neighbor hopping and an infinite annihilation rate is completely determined by the dynamics of a single random walker [Eqs. (7) and (8)] in a dual hopping environment [Eq. (6)]. This exact result allows for the exact calculation of the density in specific environments, but can also be used for extremely accurate numerical calculations of the density for any fixed inhomogeneous hopping environment or for disordered systems with subdiffusive logarithmic behavior. The approach has a straightforward extension to the calculation of correlation functions.

(iv) For disordered environments which are on average translationally and reflection invariant, and which lead to an asymptotic factorization of the two-particle conditional prob-

abilities, the density at time t with a random initial state is equal to the return probability (14) of a single particle. An important open problem is the derivation of conditions on disorder distributions under which factorization holds. For such distributions, Eq. (14) is an exact asymptotic result, and, as we would like to point out, consistent with the Smoluchovsky approach if the return probability is proportional to $1/\sqrt{D_{\mathrm{eff}}t}$. This may be a hint as to under which circumstances the Smoluchovsky treatment is adequate for the calculation of the density. Since at late times particles are separated (on average) by a distance $1/\rho\to\infty$, one would not expect a finite reaction rate or short-range interactions between particles to change this asymptotic behavior. It is interesting to note that Eq. (14) is also consistent with the renormalization group treatment of DLPA with disorder [13]. Therefore we are confident that Eq. (14) holds for a broad class of disorder distributions.

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