

**ANOMALOUS TRANSPORT PROCESSES IN  
CHEMICALLY ACTIVE RANDOM ENVIRONMENT<sup>1</sup>**

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The effect of random velocity field on the kinetics of single-species and two-species annihilation reactions is analysed near two dimensions in the framework of the field-theoretic renormalisation group. Fluctuations of particle density are modeled within the approach of Doi. The random incompressible velocity field is generated by stochastically forced Navier-Stokes equation in which thermal fluctuations—relevant below two dimensions—are taken into account.

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### 1 Introduction

The effect of density fluctuations on the asymptotics of reaction rates in low dimensionalities has attracted considerable attention recently [1,2]. Reaction rates may also be affected by fluctuations of an advective velocity field. Most work in this direction has been carried out for the case of quenched random drift [3–5]. Recently, the asymptotic behaviour of the unimolecular reaction  $A + A \rightarrow \emptyset$  in a dynamically generated random drift has been analysed with the aid of field-theoretic renormalisation group (RG) [6]. In my report I shall describe a similar approach to the bimolecular reaction  $A + B \rightarrow \emptyset$ .

Classical rate equations for densities  $n_A, n_B$  with the homogeneous initial condition  $n_A(0) = n_{A0}, n_B(0) = n_{B0}$

$$\frac{dn_A}{dt} = -K_0 n_A n_B, \quad \frac{dn_B}{dt} = -K_0 n_A n_B \quad (1)$$

yield the “normal” decay laws. Let, for definiteness,  $n_{A0} < n_{B0}$ , then

$$n_A(t) \underset{t \rightarrow \infty}{\sim} (n_{B0} - n_{A0}) \frac{n_{A0}}{n_{B0}} e^{-K_0(n_{B0} - n_{A0})t}, \quad n_B(t) \underset{t \rightarrow \infty}{\sim} (n_{B0} - n_{A0}). \quad (2)$$

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In case of equal initial densities  $n_{A0} = n_{B0} = n_0$  a powerlike decay takes over:

$$n_A(t) = n_B(t) \underset{t \rightarrow \infty}{\sim} \frac{1}{K_0 t}. \quad (3)$$

A heuristic account of initial-density fluctuations amplified by diffusion together with numerical simulations [7, 8] yield anomalous decay at  $d < 4$ , e.g. for  $n_{A0} = n_{B0} = n_0$ :

$$n_A(t) \underset{t \rightarrow \infty}{\sim} \frac{cn_0^{1/2}}{(Dt)^{d/4}}, \quad (4)$$

which is slower than the ‘‘classical’’ decay rate (3).

## 2 Second quantization for reactions

For a systematic analysis of the effect of density fluctuations it is convenient to use a field-theoretic approach. There are two possibilities available for this. First, the more widely known Martin-Siggia-Rose (MSR) approach [9], which corresponds to the solution of a Langevin-type stochastic equation to describe fluctuations. Physically, this approach is well-suited to situations in which fluctuations are of ‘‘external’’ origin, e.g. small-scale thermal fluctuations from the point of view of macroscopic (hydrodynamic) scale physics, or fluctuations caused by some external random source. Second, the several times reinvented Doi approach [10], in which the randomness is described by a probability distribution functional (PDF) or, if you like, an infinite set of probability distribution functions on a lattice, with the subsequent set of master equations instead of Langevin equations. This approach is better suited to cases, in which fluctuations of intrinsic origin are dealt with. This is the case, for instance, for density fluctuations due to randomness in the (chemical) reaction process itself.

Therefore, I will use here the Doi approach, which allows for a ‘‘mesoscopic’’ analysis of density fluctuations. To calculate expectation values with the probability distribution functional (PDF)  $P(\{n_A(\mathbf{x})\}; \{n_B(\mathbf{x})\}, t)$  for the particle densities  $n_A(\mathbf{x})$  and  $n_B(\mathbf{x})$ , the formal solution of the set of master equations for the PDF may be expressed in a functional form with the aid of bosonic field operators [10, 11] with the commutation relations

$$[\psi_A(\mathbf{x}), \psi_A^\dagger(\mathbf{x}')] = \delta(\mathbf{x} - \mathbf{x}'), \quad [\psi_A(\mathbf{x}), \psi_A(\mathbf{x}')] = [\psi_A^\dagger(\mathbf{x}), \psi_A^\dagger(\mathbf{x}')] = 0,$$

and a similar set for the  $B$  particles. In a fairly standard-looking second-quantization setting the average of an observable  $O$  may be written as a vacuum expectation value

$$\begin{aligned} \langle O(t) \rangle &= \sum_{\{n_i(\mathbf{x})\}} O[\{n_A\}, \{n_B\}] P(\{n_A\}; \{n_B\}, t) \\ &= \langle 0 | O[(\psi_A^\dagger + 1)\psi_A, (\psi_B^\dagger + 1)\psi_B] e^{-\hat{H}'t} \\ &\times e^{\int d\mathbf{x} (n_{A0}\psi_A^\dagger + n_{B0}\psi_B^\dagger - r_0\sqrt{n_{A0}n_{B0}}\psi_A^\dagger\psi_B^\dagger)} | 0 \rangle, \end{aligned} \quad (5)$$

with the kinetic operator

$$\begin{aligned} \hat{H}' = & \int d\mathbf{x} \left\{ \psi_A^+ \nabla(\mathbf{v}\psi_A) + \psi_B^+ \nabla(\mathbf{v}\psi_B) - D_{A0}\psi_A^+ \nabla^2 \psi_A - D_{B0}\psi_B^+ \nabla^2 \psi_B \right. \\ & \left. + K_0 (\psi_A^+ + \psi_B^+ + \psi_A^+ \psi_B^+) \psi_A \psi_B \right\}. \end{aligned} \quad (6)$$

The last exponential in (5) corresponds to the initial PDF. A customary choice is the Poisson distribution for the local particle number [1,2]. However, in view of the hostile nature of interaction I have allowed for negative initial correlations by using in (5) a *bivariate* Poisson distribution [12]. This choice leads to the following expressions for the low-order moments of initial densities:

$$\begin{aligned} \overline{n_A(\mathbf{x}, 0)} = n_{A0}, \quad \overline{n_B(\mathbf{x}, 0)} = n_{B0}, \quad \overline{\Delta n_A(\mathbf{x}, 0) \Delta n_B(\mathbf{x}', 0)} = -r_0 \sqrt{n_{A0} n_{B0}} \delta_{\mathbf{x}, \mathbf{x}'}, \\ \overline{\Delta n_A(\mathbf{x}, 0) \Delta n_A(\mathbf{x}', 0)} = n_{A0} \delta_{\mathbf{x}, \mathbf{x}'}, \quad \overline{\Delta n_B(\mathbf{x}, 0) \Delta n_B(\mathbf{x}', 0)} = n_{B0} \delta_{\mathbf{x}, \mathbf{x}'}. \end{aligned} \quad (7)$$

Physically this corresponds to thermal fluctuations with anticorrelations in initial reactant densities.

### 3 Dynamic action for the advection-diffusion-controlled reaction $A + B \rightarrow \emptyset$

Construction of perturbation theory through the  $T$  exponent for the evolution operator

$$U(t, t_0) = e^{\hat{H}'_0 t} e^{-\hat{H}'(t-t_0)} e^{-\hat{H}'_0 t_0} = T e^{-\int_{t_0}^t \hat{H}'_t dt}$$

allows to write the expectation value (5) as the following functional integral:

$$\langle O(t) \rangle = \int \mathcal{D}[\psi] O_N(1, \psi_A, 1, \psi_B) e^{S_0 + S_1}, \quad (8)$$

where  $O_N$  is the normal symbol of the operator  $O$

$$O[\psi_A^+ \psi_A, \psi_B^+ \psi_B] = N[O_N(\psi_A^+, \psi_A, \psi_B^+, \psi_B)],$$

$S_1$  is the dynamic action [2]

$$\begin{aligned} S_1 = & - \int d\mathbf{x} dt \left\{ \psi_A^+ \partial_t \psi_A + \psi_B^+ \partial_t \psi_B + \psi_A^+ \nabla(\mathbf{v}\psi_A) + \psi_B^+ \nabla(\mathbf{v}\psi_B) \right. \\ & \left. - D_{A0}\psi_A^+ \nabla^2 \psi_A - D_{B0}\psi_B^+ \nabla^2 \psi_B + K_0 (\psi_A^+ + \psi_B^+ + \psi_A^+ \psi_B^+) \psi_A \psi_B \right\} \end{aligned} \quad (9)$$

and  $S_0$  contains terms brought about by the initial bivariate Poisson distribution

$$S_0 = \int d\mathbf{x} \left[ n_{A0}\psi_A^+ + n_{B0}\psi_B^+ - r_0 \sqrt{n_{A0} n_{B0}} \psi_A^+ \psi_B^+ \right].$$

Schwinger equations with respect to  $\psi_A^+, \psi_B^+$

$$\langle \partial_t \psi_A + \nabla(\mathbf{v}\psi_A) - D_{A0}\nabla^2 \psi_A + K_0 \psi_A \psi_B \rangle = n_{A0} \delta(t), \quad (10)$$

$$\langle \partial_t \psi_B + \nabla(\mathbf{v}\psi_B) - D_{B0}\nabla^2 \psi_B + K_0 \psi_A \psi_B \rangle = n_{B0} \delta(t), \quad (11)$$

in the mean-field approximation yield the classical rate equations (1) for the homogeneous average densities  $\langle n_A(t) \rangle = \langle \psi_A(t) \rangle$ ,  $\langle n_B(t) \rangle = \langle \psi_B(t) \rangle$ . It should be borne in mind, however, that the second and higher order moments of the fields  $\psi_A$  and  $\psi_B$  are not equal to the corresponding moments of the densities. For instance, the pair correlations of  $A$  particles are given by  $\langle n_A(t, \mathbf{x}) n_A(t, \mathbf{x}') \rangle = \langle [\psi_A(t, \mathbf{x}) \psi_A(t, \mathbf{x}') + \delta(\mathbf{x} - \mathbf{x}') \psi_A(t, \mathbf{x})] \rangle$ .

To describe fluctuations of the drift field  $\mathbf{v}$  in (9) I use random velocity field generated by the transverse stochastic Navier-Stokes equation

$$\partial_t \mathbf{v} + P(\mathbf{v} \cdot \nabla) \mathbf{v} - \nu_0 \nabla^2 \mathbf{v} = \mathbf{f}^v \quad (12)$$

with the incompressibility conditions:  $\nabla \cdot \mathbf{v} = 0$ ,  $\nabla \cdot \mathbf{f}^v = 0$ . For the random force the Gaussian distribution with zero mean and the correlation function

$$\langle f_m^v(\mathbf{x}_1, t_1) f_n^v(\mathbf{x}_2, t_2) \rangle = \delta(t_1 - t_2) \int \frac{d\mathbf{k}}{(2\pi)^d} P_{mn}(\mathbf{k}) d_f(k) e^{i\mathbf{k} \cdot (\mathbf{x}_1 - \mathbf{x}_2)} \quad (13)$$

is assumed. In (13)  $P_{mn}(\mathbf{k}) = \delta_{mn} - k_m k_n / k^2$  is the transverse projection operator in the wave-vector space, and  $d_f(k)$  is a function of the wave number  $k$  and the parameters of energy pumping, which is used to produce stationary random drift. The kernel function is often chosen in the nonlocal form

$$d_f(k) = g_{10} \nu_0^3 k^{4-d-2\epsilon} \quad (14)$$

to generate turbulent velocity field with Kolmogorov's scaling [13, 14] (which is achieved by choosing  $\epsilon = 2$ ).

The stochastic Navier-Stokes equation (12) is a Langevin-type equation leading to a MSR operator functional which in the functional-integral form gives rise to the following action functional

$$\begin{aligned} S_2 &= \frac{1}{2} \int dt dx dx' \tilde{\mathbf{v}}(\mathbf{x}, t) \cdot \tilde{\mathbf{v}}(\mathbf{x}', t) d_f(|\mathbf{x} - \mathbf{x}'|) \\ &+ \int dt dx \tilde{\mathbf{v}} \cdot [-\partial_t \mathbf{v} - (\mathbf{v} \cdot \nabla) \mathbf{v} + \nu_0 \nabla^2 \mathbf{v}]. \end{aligned} \quad (15)$$

Here, transverse auxiliary vector field  $\tilde{\mathbf{v}}$  is implied, and therefore the projection operator  $P$  from (12) and (13) has been omitted.

Combined averaging over density and velocity fluctuations yields

$$\langle O(t) \rangle = \int \mathcal{D}[\psi, \mathbf{v}] O_N(1, \psi_A, 1, \psi_B) e^{S_0 + S_1 + S_2} \quad (16)$$

for the expectation value of the observable  $O$ .

#### 4 Decay asymptotics controlled by stable fixed points

Power counting shows that in the case in which all three reaction terms in (9) have the same scaling dimension the critical dimension of the model is two [2]. Near two dimensions, however, the drift part (15) of the dynamic action with the nonlocal kernel (14) is not multiplicatively

renormalisable. Therefore I have used the kernel function [15] with a local term added at the outset:

$$d_f(k) = g_{10}\nu_0^3 k^{4-d-2\epsilon} + g_{20}\nu_0^3 k^2. \quad (17)$$

Apart from rendering the field theory multiplicatively renormalisable, the local term also has an important physical meaning: with a suitable choice of the parameter  $g_{20}$  it describes thermal fluctuations of the velocity field near equilibrium.

Taking this into account, I write the renormalised action in the form

$$\begin{aligned} S = & - \int d\mathbf{x} dt \left\{ \psi_A^\dagger \partial_t \psi_A + \psi_B^\dagger \partial_t \psi_B + \psi_A^\dagger \nabla(\mathbf{v}\psi_A) + \psi_B^\dagger \nabla(\mathbf{v}\psi_B) \right. \\ & - u_{A\nu} Z_{2A} \psi_A^\dagger \nabla^2 \psi_A - u_{B\nu} Z_{2B} \psi_B^\dagger \nabla^2 \psi_B \\ & + \lambda \nu \mu^{-2\delta} Z_4 (\psi_A^\dagger + \psi_B^\dagger + \psi_A^\dagger \psi_B^\dagger) \psi_A \psi_B \\ & - \frac{1}{2} \tilde{\mathbf{v}} \left[ g_1 \nu^3 \mu^{2\epsilon} (-\nabla^2)^{1-\delta-\epsilon} - g_2 \nu^3 \mu^{-2\delta} Z_3 \nabla^2 \right] \tilde{\mathbf{v}} \\ & + \tilde{\mathbf{v}} \cdot \left[ \partial_t \mathbf{v} + (\mathbf{v} \cdot \nabla) \mathbf{v} - \nu Z_1 \nabla^2 \mathbf{v} \right] \left. \right\} \\ & + \int d\mathbf{x} \left[ \kappa_A \mu^d Z_{5A} \psi_A^\dagger + \kappa_B \mu^d Z_{5B} \psi_B^\dagger - \rho \mu^d Z_5 \psi_A^\dagger \psi_B^\dagger \right], \end{aligned} \quad (18)$$

in which, apart from the standard renormalisation of the dynamic action, also the renormalisation of the initial condition—predicted by power counting and confirmed by calculations—is introduced.

Renormalisation constants have been calculated in one-loop approximation with the use of combined dimensional and analytic regularisation with the parameters  $\epsilon$  and  $\delta = (d-2)/2$ , which eventually give rise to a two-parameter expansion of critical exponents and other physical quantities.

The unrenormalised parameters of the initial conditions have positive canonical scaling dimensions. This means that the corresponding running parameters grow in the long-time large-scale limit. Therefore, some kind of partial summation of the perturbative expansion is called for to cope with this problem. A natural way would be the use of skeleton equations for Green functions with dressed field averages and correlation functions. In the case of single-species annihilation reaction  $A + A \rightarrow \emptyset$  this leads to well-controlled estimates of the behaviour of scaling functions in the long-time limit [1]. Basically, this amounts to independence of the asymptotics of the initial density [16]. In the present case of bimolecular annihilation, however, a similar direct summation has not been found [2]. Unfortunately, I have not been able to do any better with an analytic solution of the set of integro-differential equations, which can be written for the dressed one-point and two-point Green functions of the present model and incorporate the effect of initial conditions completely.

In the leading order in the coupling constant  $\lambda$  the initial density fluctuations change the classical rate eqs (1) by the addition of an inhomogeneous term. This leads to the system

$$\begin{aligned} \partial_t n_A &= -\lambda \nu \mu^{-2\delta} \left\{ n_A n_B - \frac{\rho \mu^d}{[4\pi\nu(u_A + u_B)t]^{d/2}} \right\}, \\ \partial_t n_B &= -\lambda \nu \mu^{-2\delta} \left\{ n_A n_B - \frac{\rho \mu^d}{[4\pi\nu(u_A + u_B)t]^{d/2}} \right\}, \end{aligned} \quad (19)$$

with the initial condition:  $n_A(0) = \kappa_A \mu^d Z_{5A}$ ,  $n_B(0) = \kappa_B \mu^d Z_{5B}$ . For the important special case  $n_A = n_B = n$  the special Riccati's equation results. The solution is known and may be expressed in terms of modified Bessel functions  $K_{2/(4-d)}$  and  $I_{2/(4-d)}$ . In this solution the blow-up of initial conditions is controllable and the asymptotic behaviour of the particle density may be inferred. However, the effect of initial conditions is not fully accounted for by the manageable system of eqs (19), and therefore the following results give the correct asymptotic density decay with this provision only.

The renormalised action (18) gives rise to a system of characteristic equations with four IR stable physical fixed points with the following asymptotic decay of the density.

(i) Gaussian fixed point

$$g_1^* = g_2^* = \lambda^* = 0.$$

The Gaussian fixed point is stable, when

$\epsilon < 0$ ,  $\delta > 0$ . Asymptotic decay of the number density in terms of physical (unrenormalised) parameters

$$n(t) \underset{t \rightarrow \infty}{\sim} \frac{\sqrt{r_0 n_0}}{[4\pi(D_{A0} + D_{B0})t]^{d/4}} \quad (20)$$

is not given by the classical solution (3) but is slower. This is different from the unimolecular case [6] in which at the Gaussian fixed point the mean-field solution holds. Note that there is no dependence on the rate coefficient in (20), but the parameter of initial correlations remains. The influence of initial correlations becomes irrelevant and the mean-field decay  $\propto 1/t$  is restored only at  $d > 4$  [2].

(ii) Thermal fixed point

$$g_1^* = 0, \quad g_2^* = -32\pi\delta, \quad u^* = \frac{\sqrt{17}-1}{2}, \quad \lambda^* = -2\pi(\sqrt{17}-1)\delta.$$

The basin of attraction of this fixed point is  $\delta < 0$ ,  $2\epsilon + 3\delta < 0$ . Decay rate is faster than the initial-density-fluctuation induced:

$$n(t) \underset{t \rightarrow \infty}{\sim} \frac{\sqrt{r_0 n_0}}{[4\pi\nu_0(\sqrt{17}-1)\tau]^{d/4}} \left(\frac{\tau}{t}\right)^{1/2}. \quad (21)$$

Here,  $\tau$  is a reference time scale and  $\delta = d/2 - 1$ .

(iii) Reactive kinetic fixed point

$$g_1^* = \frac{64\pi}{9} \frac{\epsilon(2\epsilon + 3\delta)}{\epsilon + \delta}, \quad g_2^* = \frac{64\pi}{9} \frac{\epsilon^2}{\delta + \epsilon},$$

$$u^* = \frac{\sqrt{17}-1}{2}, \quad \lambda^* = -\frac{4\pi}{3}(\sqrt{17}-1)(\epsilon + 3\delta),$$

is stable, when  $\epsilon > 0$ ,  $-\frac{2}{3}\epsilon < \delta < -\frac{1}{3}\epsilon$ . To linear order in  $\delta$ ,  $\epsilon$ , decay exponent the same as in thermal fixed point

$$n(t) \underset{t \rightarrow \infty}{\sim} \frac{\sqrt{r_0 n_0}}{[4\pi\nu_0(\sqrt{17}-1)\tau]^{d/4}} \left(\frac{\tau}{t}\right)^{1/2}. \quad (22)$$

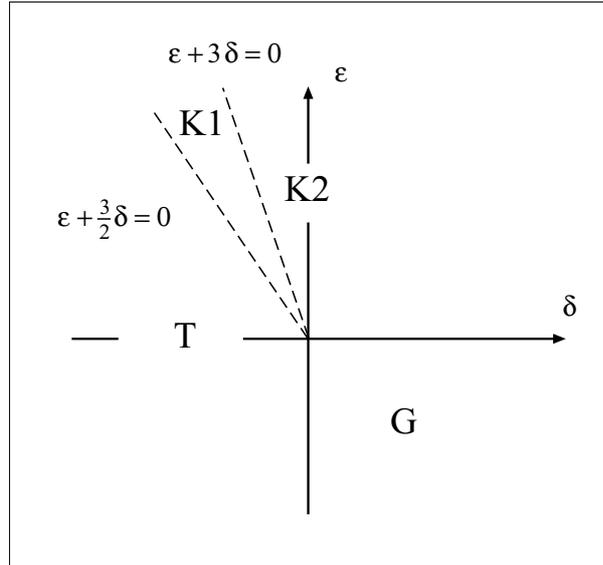


Fig. 1. Basins of attraction of the Gaussian fixed point (G), thermal fixed point (T), the reactive kinetic fixed (K1) and the passive kinetic fixed point (K2) in the  $(\delta, \epsilon)$  plane.

The independence of the exponent of time of  $\delta$  and  $\epsilon$  in (21) and (22) is most probably an artifact of the one-loop approximation.

(iv) Passive kinetic fixed point

$$g_1^* = \frac{64\pi}{9} \frac{\epsilon(2\epsilon + 3\delta)}{\epsilon + \delta}, \quad g_2^* = \frac{64\pi}{9} \frac{\epsilon^2}{\delta + \epsilon}, \quad u^* = \frac{\sqrt{17} - 1}{2}, \quad \lambda^* = 0,$$

is stable, when  $\epsilon > 0, \delta > -\frac{1}{3}\epsilon$ . Decay rate is faster than the initial-density-fluctuation induced here, too:

$$n(t) \underset{t \rightarrow \infty}{\sim} \frac{\sqrt{r_0 n_0}}{[4\pi\nu_0(\sqrt{17} - 1)\tau]^{d/4}} \left(\frac{\tau}{t}\right)^{d/4(1-\epsilon/3)}. \quad (23)$$

Here,  $\tau$  is the reference time scale. The decay exponent in (23) is exact.

From these results it follows that the decay exponent is a continuous function of  $\delta = d/2 - 1$  and  $\epsilon$ —apart from logarithmic corrections on the basin boundaries.

As in the case of unimolecular reaction [6], the fixed point corresponding to the pure diffusion-limited reaction

$$g_1^* = g_2^* = 0, \quad \lambda^* = -4\pi(\sqrt{17} - 1)\delta$$

is unstable in  $d < 2$ . This means that any velocity fluctuations (including the ubiquitous thermal noise!) drive the system to the advection-diffusion-controlled regime with different decay exponents.

In case of unequal initial densities the system (19) leads to a particular form of Riccati's general equation, for which the solution seems to be unknown. Therefore, I will not discuss this case here in the hope that a solution of this equation in terms of known special functions may eventually be found with a reasonable effort.

## 5 Conclusions

The main physical result of this work is that diffusion-limited two-species annihilation reaction is shown to be unstable to short-range (thermal) velocity fluctuations for  $d \leq d_c = 2$  and unstable to long-range (turbulent) velocity fluctuations for  $d \geq 2$ . Decay exponents in four stable advection-diffusion-controlled regimes have been calculated at one-loop order. Although the renormalisation and fixed-point analysis are fairly straightforward, the asymptotic analysis of scaling functions is not. The blow-up of the effective (running) initial conditions in the scaling functions is difficult to control, in contrast with the single-species case, and a firm conclusion about the asymptotics of scaling functions is not yet available.

As to possible generalisations, it would be interesting to amend the decay analysis by a similar treatment of a stationary state with reactant sources.

It seems quite plausible that the asymptotic behaviour of the density heavily depends on the localisation of the initial density profiles; therefore, an analysis of the problem with localised initial conditions would be desirable.

Due to the incompressibility condition imposed on the drift field, the present results have a direct physical meaning at  $d \geq 2$  only. Thus, the effect of compressibility should be analysed. This, however, does not seem to be feasible at present in the full stochastic Navier-Stokes framework. Therefore, to make some progress in this direction, it would be interesting to analyse the effect of velocity fluctuations with given statistics instead of dynamically generated random drift.

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