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Selection of length distributions in living polymers

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The steady state distribution of polymer (or micelle) lengths under nonequilbrium conditions in which monomers are continuously extracted from a system is studied. The dynamical equations describing this process exhibit a one-parameter family of steady state distributions. A study of the dynamical equations suggests that they exhibit either a linear marginal or nonlinear marginal selection, depending on the control parameters of the model. The selection is explicitly demonstrated for a simplified linear version of the dynamical equations.

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Selection of patterns in systems far from thermal equilibrium has been extensively studied in recent years. Most studies have been concerned with a situation where a stable state (usually nonuniform) of a dissipative system propagates into an initially (usually uniform) unstable region [1–7]. Experimentally, this may be realized when a stable system is abruptly brought above its stability limit. A small perturbation may then locally drive the system into a new stable state, which eventually spreads into the rest of the space. Commonly studied systems are Rayleigh-Benard convection cells, Taylor instabilities, chemical reactions with diffusion, solidification fronts, and others. Usually, these systems possess a band of linearly stable nonuniform states. Questions which one would like to answer are the nature of the nonuniform pattern of the resulting stable state, the mechanism by which this pattern is selected, and the speed with which the front separating the two phases is moving.

A simple model for which the selection mechanism can be demonstrated was analyzed by Aronson and Weinberger [1]. They considered a nonlinear diffusion equation of the form

$$\frac{\partial\phi}{\partial t}=\frac{\partial^2\phi}{\partial x^2}+f(\phi)\ , \tag{1}$$

where $f(\phi)$ is chosen such that (1) has a stable uniform state at, say, $\phi=1$ and an unstable state at $\phi=0$. A front separating the $\phi=1$ state from the $\phi=0$ one propagates into the unstable phase with a velocity v. Steady state front solutions of Eq. (1) with any velocity v may be found. However, it has been shown that for a large class of functions $f(\phi)$ and for physically relevant (namely, localized) initial conditions, a particular velocity v is selected. Depending on the function $f(\phi)$, the selected velocity may either correspond to a marginally stable fixed point or to a different fixed point which is referred to as case II (or nonlinear) marginal stability [1,3,6]. These results were subsequently generalized and applied to more complicated situations in which the stable state is nonuniform.

In the present work we consider a different class of systems in which selection may take place. Here we do not study front propagation into an unstable phase, but rather the steady state distribution of certain systems away from thermal equilibrium. Since these systems are locally stable one may, in principle, study the effect of random noise on the selection mechanism in this type of system. This class may also provide a simple example in which both marginal stability and case II marginal stability may take place. We consider a system of aggregates (say, polymers or micelles) each composed of n basic units. Let c_n be the number of n-mers in the system. Under equilibrium conditions the system reaches a well defined length distribution c_n. We now drive the system out of equilibrium by pulling out monomers (or small size molecules) at some given rate. This could be achieved experimentally, say, by providing a surface on which monomers could be adsorbed, thus leaving the system. Clearly, under these conditions the c_n's decrease with time, and the system eventually vanishes. However, one may look at the concentration of n-mers $x_n\equiv c_n/\overline{c}$, with

$$\overline{c}=\sum_{n=1}^{\infty} nc_n\ ,$$

and ask whether these quantities reach a well defined limit for large time t. We show that under the nonequilibrium conditions specified above, the system has a one parameter family of linearly stable steady state distributions x_n. We then consider the selection mechanism by which a particular distribution is selected. The system is found to exhibit either a marginal stability or nonmarginal stability selection, depending on the parameters which define its dynamics. The time evolution of these systems is similar to that of two dimensional soap froth, which has recently been studied in detail [8]. Soap froth (and other cellular structures such as polycrystalline films) are composed of $n \geq 3$ sided cells which display dynamics where $n = 3,4,5$ sided cells tend to decrease in size with time and eventually disappear. A selection mechanism governing the long time evolution of these systems has recently been discussed [9]. In the following we introduce a model for the dynamics of interacting polymers and demonstrate that it has a one parameter family of fixed point distributions. The dynamics of the model is studied numerically, and the selection mechanisms are examined. A simplified linear model is then considered. The model is exactly soluble, and the selection mechanism can be demonstrated analytically. The mathematical details of this analysis are given in the Appendix.

We now introduce a model for the dynamics of aggregates (such as polymers or micelles). For simplicity, it is assumed that the interaction between the various n-mers takes place via an association-dissociation process in which an n-mer absorbs or emits a single monomer and becomes an $(n+1)$ or $(n-1)$-mer, respectively. The equations which govern this process take the form

$$\frac{dc_1}{dt} = -kx_1 \sum_{n=1}^{\infty} c_n + \bar{k} \sum_{n=2}^{\infty} c_n - ac_1 \ ,$$
$$\frac{dc_2}{dt} = kx_1(c_1/2 - c_2) + \bar{k}(c_3 - c_2/2) \ , \tag{2}$$
$$\frac{dc_n}{dt} = kx_1(c_{n-1} - c_n) + \bar{k}(c_{n+1} - c_n) \quad \text{for } n \geq 3 \ .$$

Here $\bar{k}$ is the dissociation rate, kx_1 is the association rate between an n-mer and a monomer, and a is the rate at which monomers are pulled out of the system. For simplicity we assume that k and $\bar{k}$ are independent of n. Note that the equation for c_2 is slightly different from those for c_n, $n \geq 3$. First, the rate at which two monomers combine to yield a dimer is $kx_1/2$, and not kx_1. In addition, a dissociation process in a dimer may take place at a single bond rather than two (those close to the two edges) for $n \geq 3$. The dissociation rate is thus $\bar{k}/2$ rather than $\bar{k}$. Summing Eqs. (2) one finds

$$\frac{d\bar{c}}{dt} = -ac_1 \ , \tag{3}$$

which gives the rate at which the number of units in the system decreases in time. System (2) is equivalent to the following equations for the concentrations x_n:

$$\frac{dx_1}{dt} = -kx_1 \sum_{n=1}^{\infty} x_n + \bar{k} \sum_{n=2}^{\infty} x_n - ax_1 + ax_1^2 \ ,$$
$$\frac{dx_2}{dt} = kx_1(x_1/2 - x_2) + \bar{k}(x_3 - x_2/2) + ax_1x_2 \ , \tag{4}$$
$$\frac{dx_n}{dt} = kx_1(x_{n-1} - x_n) + \bar{k}(x_{n+1} - x_n) + ax_1x_n \quad \text{for } n \geq 3 \ .$$

This is a set of nonlinear equations with nonlocal interactions, where the "head" of the distribution, x_1, directly interacts with its "tail," x_n, for arbitrarily large n. The x_n distribution corresponding to the fixed points of these equations together with Eq. (3) yield the long time behavior of c_n. Note that system (4) preserves the normalization condition

$$\sum_{n=1}^{\infty} nx_n = 1 \ . \tag{5}$$

More precisely, summing Eqs. (4) gives

$$\frac{d}{dt} \sum_{n=1}^{\infty} nx_n = ax_1 \left[\sum_{n=1}^{\infty} nx_n - 1 \right] \ .$$

By examining Eqs. (4) one finds that in general they do not have just one fixed point, but rather a one parameter family of fixed point distributions, x_n. To see that this indeed is the case, one first makes an arbitrary choice for x_1 and x_2. Then the second equation of system (4) determines x_3. In a similar way we can successively solve the remaining equations, obtaining x_n for arbitrary $n > 3$. The two free parameters x_1 and x_2 may now be varied so as to satisfy the equation for x_1 [or, equivalently, the normalization condition (5)], leaving one free parameter. As long as the x_n's obtained by this procedure are non-negative, the resulting distribution is physically relevant. These distributions may, for example, be parametrized by x_1.

To evaluate the fixed point distributions one notes that, for a given x_1, Eq. (4) for $n \geq 3$ is a second order difference equation with constant coefficients. Thus it has a solution of the form

$$x_n = A\lambda_-^{n-2} + B\lambda_+^{n-2} \ , \quad n \geq 2 \ , \tag{6}$$

where $\lambda_\pm$ are the roots of the characteristic equation

$$\bar{k}\lambda^2 - (\bar{k} + kx_1 - ax_1)\lambda + kx_1 = 0 \ . \tag{7}$$

Here A and B are parameters which are determined by the first two equations in (4),

$$\frac{A}{1-\lambda_-} + \frac{B}{1-\lambda_+} = \frac{(k-a)x_1^2 + ax_1}{\bar{k} - kx_1} \ ,$$
$$A[\bar{k}\lambda_- - (\bar{k}/2 + kx_1 - ax_1)] + B[\bar{k}\lambda_+ - (\bar{k}/2 + kx_1 - ax_1)] = -kx_1^2/2 \ . \tag{8}$$

Solving Eq. (8) for A and B one obtains a fixed point distribution for any x_1. However, for the distribution to be physical, all x_n's have to be non-negative. This is the case provided (a) the roots $\lambda_\pm$ are real with $|\lambda_-| < \lambda_+ < 1$, (b) $A + B \geq 0$, and (c) $B \geq 0$. The first condition ensures that the solution is not oscillating but purely exponentially decaying near infinity. The second condition is just $x_2 \geq 0$. The last condition is required since B corresponds to the dominant part of the solution as $n \to \infty$. A negative B would imply that x_n becomes negative for sufficiently large n.

Examining Eq. (7) one finds that there is $x_M > 0$ such that $\lambda_\pm$ satisfy condition (a) provided $0<x_1<x_M$. Solving Eq. (8) for A and B, one verifies that condition (b) is always satisfied if $0<x_1<x_M$. Regarding condition (c) two possible types of behavior are found, depending on the parameters k and a which define the model (the third parameter $\bar{k}$ may be set to 1. It determines the time scale in the problem and does not affect the fixed-point distributions). The parameter B either satisfies $B \geq 0$ for $0<x_1<x_M$, in which case all fixed points with $0<x_1<x_M$ are physical [see Fig. 1(a)], or there exists $0<x_S<x_M$ such that $B \geq 0$ for $0<x_1<x_S$, while B changes sign at $x_1=x_S$ [see Fig. 1(b)]. In this case only the $0<x_1 \leq x_S$ are physically relevant. Following the work of Aronson and Weinberger [1], and Dee, Langer, and Muller-Krumbhaar [2] one may conjecture that in the first case the marginal fixed point corresponding to $x_1=x_M$ is selected, in the sense that as long as the initial length distribution decays sufficiently fast with n, the system evolves towards the $x_1=x_M$ fixed point. On the other hand, in the second case (the nonlinear marginal stability, or case II) the selected fixed point is the one corresponding to $x_1=x_S$. In both cases the selected distribution corresponds to the physically accessible fixed point with the fastest decay rate of x_n with n. Numerical integration of Eq. (4) shows that indeed the long time behavior of the system is governed by either the $x_1=x_M$ or the $x_1=x_S$ fixed points depending on the behavior of B. Examples of some distributions are given in Fig. 2. The (a,k) phase diagram is given in Fig. 3. The phase diagram exhibits a line separating two regions, one in which a marginal selection (M) takes place, and the other in which nonlinear marginal selection (S) is valid.

In order to study the evolution of this system more closely, we introduce a continuum model corresponding to the dynamics of aggregates. Let the length parameter, n, be replaced by a continuous variable $x \geq 0$, and denote the concentration of polymers of length between x and $x+dx$ at time t by $\phi(x,t)dx$. The concentration function $\phi(x,t)$ satisfies an equation of the form

$$\frac{\partial \phi}{\partial t}=a_2\frac{\partial^2 \phi}{\partial x^2}+a_1\frac{\partial \phi}{\partial x}+a_0\phi \ , \tag{9}$$

for $x>0$ where a_2, a_1, and a_0 are linear functions of $\phi(0,t)$. Equation (9) should be supplemented by the normalization condition

$$\int_0^\infty x\phi(x,t)dx=1 \ . \tag{10}$$

This is a nonlinear differential equation with long range interactions, where $\phi(0,t)$ interacts directly with $\phi(x,t)$ for any $x \geq 0$.

In trying to establish the selection mechanism for Eq. (9) with (10), one notes that due to the normalization condition, one cannot apply the methods of Aronson and Weinberger in this case. The reason is that any pair of

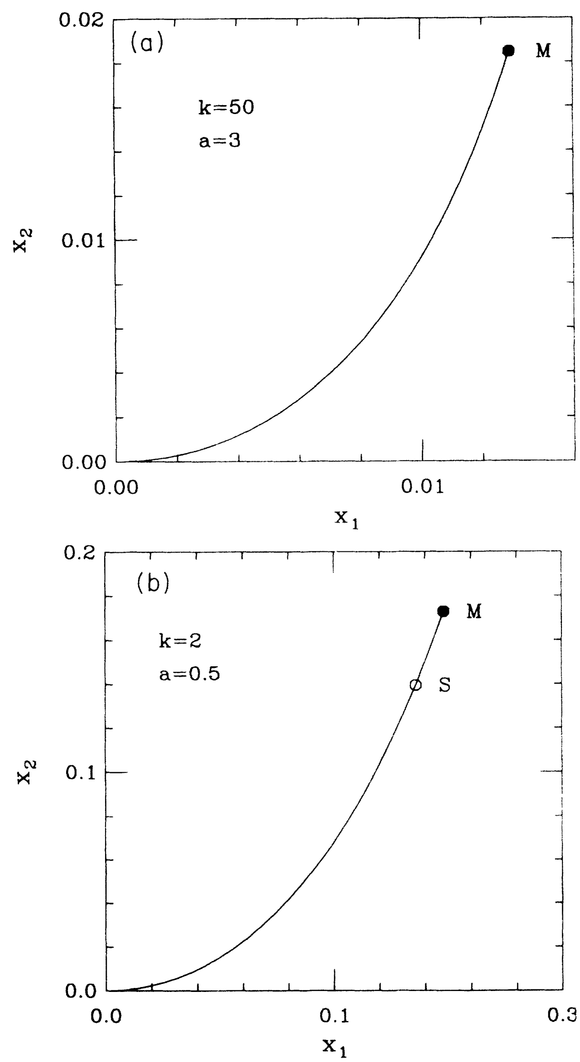

FIG. 1. A cut of the steady state distribution in the (x_1,x_2) plane: (a) in the region where a marginal fixed point M is selected and (b) in a region where nonlinear selection (S) takes place.

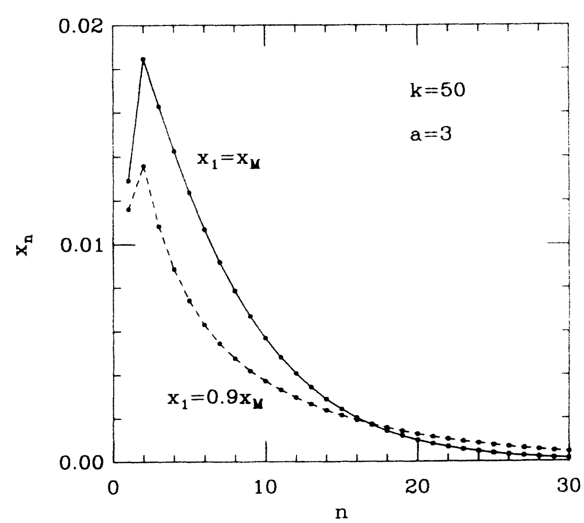

FIG. 2. An example of some steady state distributions. In this case the $x_1=x_M$ fixed point is selected.

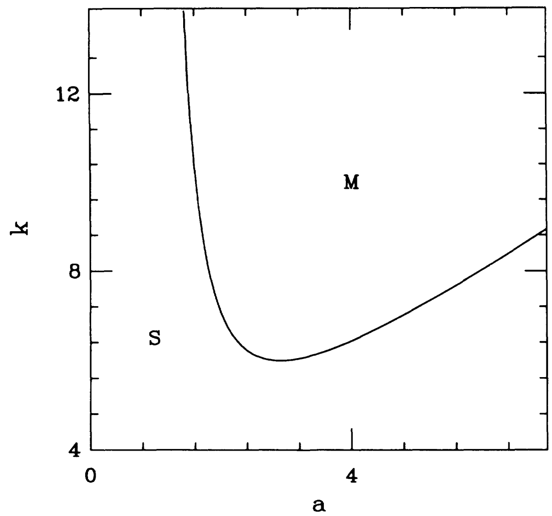

FIG. 3. The (a,k) phase diagram of the model Eq. (4). The regions M in which marginal selection is valid and S in which nonlinear marginal selection is valid are marked. $\bar{k}$ is equal to 1.

non-negative functions $\phi(x,t)$ which satisfy (10) have to intersect. Therefore one cannot simply apply the positivity condition which was used for analyzing the dynamics of front propagation governed by equations such as (1). To proceed, we assume that the coefficients a_i, $i=0,1,2$ are constants independent of $\phi(0,t)$, and study the dynamics of the resulting linear equation. This may be justified by the fact that near the fixed-point distribution $\phi(0,t)$ is independent of t and may thus be replaced by a constant. By rescaling t and x in Eq. (9) one may replace, say, a_2 and a_0 by 1 and consider the dynamics of the following equation:

$$\frac{\partial\phi}{\partial t}=\frac{\partial^2\phi}{\partial x^2}+c\frac{\partial\phi}{\partial x}+\phi \ , \tag{11}$$

where c is a constant, $\phi\geq 0$, and $x\geq 0$. This equation is supplemented by the normalization condition (10). It has a one parameter family of fixed-point distributions

$$\phi(x)=Ae^{-\alpha_+ x}+Be^{-\alpha_- x} \ , \tag{12}$$

where A and B are constants, and $\alpha_\pm$ are the roots of the quadratic equation

$$\alpha^2-c\alpha+1=0 \ . \tag{13}$$

This equation has two real roots $0<\alpha_-<\alpha_+$ as long as $c>2$, and the resulting function ϕ is non-negative provided $A+B\geq 0$ and $B\geq 0$. The normalization condition (10) implies that A and B are related via

$$\frac{A}{\alpha_+^2}+\frac{B}{\alpha_-^2}=1 \ . \tag{14}$$

Therefore, one ends up with a one parameter (say, B) family of fixed-point distributions of the form of (12). The question now is which of these distributions is selected by the dynamics of the system. In the following we demonstrate that if the initial distribution $\phi(x,0)$ decays faster than $e^{-(c/2)x}$, the selected fixed point is the one with $B=0$, namely, the one with the fastest decay tail.

To study the evolution of $\phi(x,t)$ we consider the deviation $\delta\phi(x,t)$ from the $B=0$ fixed point solution (12),

$$\phi(x,t)=\alpha_+^2 e^{-\alpha_+ x}+\delta\phi(x,t) \ , \tag{15}$$

where $\delta\phi(x,t)$ satisfies the same equation as ϕ, namely Eq. (11), with the normalization condition

$$\int_0^\infty x\,\delta\phi(x,t)dx=0 \ . \tag{16}$$

If the initial distribution $\phi(x,0)$ decays faster than $e^{-(c/2)x}$, so does $\delta\phi(x,0)$ (since $\alpha_+\geq c/2$). One therefore has to show that for such initial distributions the function $\delta\phi(x,t)$ decays to zero in time, thus selecting the $B=0$ fixed point. Before demonstrating that this indeed is the case, it is instructive to consider some simple solutions for $\delta\phi(x,t)$. Equation (11) together with (16) have solutions of the form

$$\delta\phi(x,t)=e^{\gamma t}(A_1 e^{-\alpha_1 x}+A_2 e^{-\alpha_2 x}) \ , \tag{17}$$

where for any given γ, $\alpha_{1,2}$ (with $\alpha_2\geq\alpha_1$) satisfy

$$\alpha^2-c\alpha+1=\gamma \ , \tag{18}$$

and $A_1/\alpha_1^2+A_2/\alpha_2^2=0$ (see Fig. 4). It is clear from Fig. 4 that as long as $\alpha_-\leq\alpha_1$, $\alpha_2\leq\alpha_+$, one has $\gamma<0$, and the perturbation decays to zero. On the other hand for $\alpha_2>\alpha_+$ (or, equivalently, for $\alpha_1<\alpha_-$), the time exponent satisfies $\gamma>0$ and the perturbation increases to infinity. This is consistent with the assertion that initial distributions $\phi(x,0)$ which decay sufficiently fast with x evolve in time to the $B=0$ fixed point.

To prove this point in general, one introduces $\psi(x,t)$ defined by

$$\delta\phi(x,t)=e^{-(c/2)x}e^{(1-c^2/4)t}\psi(x,t) \ . \tag{19}$$

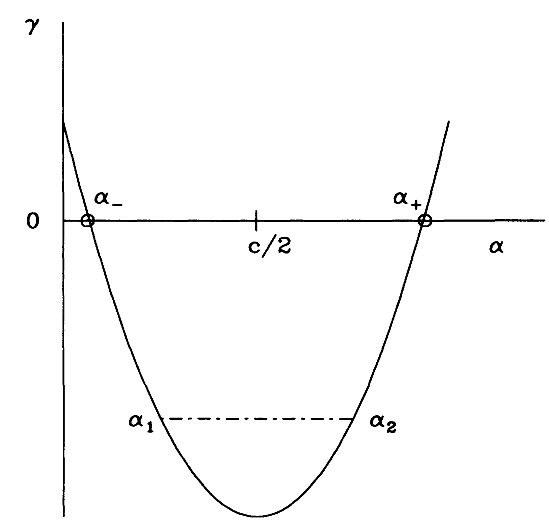

FIG. 4. The dispersion corresponding to the model Eq. (11).

It satisfies the equation

$$\frac{\partial\psi}{\partial t}=\frac{\partial^2\psi}{\partial x^2}, \tag{20}$$

with

$$\int_0^\infty x e^{-(c/2)x}\psi(x,t)dx=0 . \tag{21}$$

These equations have a set of eigenfunctions

$$e^{-k^2 t}\sin(kx-\theta_k), \quad k>0 , \tag{22}$$

where the θ_k's are chosen such that the integral condition (21) is satisfied. In the Appendix we use the eigenfunction expansion associated with (22) to show that solutions of (20) and (21) remain bounded as $t\to\infty$. Therefore, for any initial configuration $\delta\phi(x,0)$ which decays faster than $e^{-(c/2)x}$, the solution $\delta\phi(x,t)$ of (11) and (16) decays to zero at large t. The $B=0$ fixed point is thus selected.

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APPENDIX

In this appendix we show that the evolution problem given by Eqs. (20) and (21) has bounded solutions. After appropriate rescaling we rewrite this problem as

$$\frac{1}{c^2}\frac{\partial\psi_t}{\partial t}=\frac{\partial^2\psi_t}{\partial x^2}, \qquad \int_0^\infty \rho(x)\psi_t(x)dx=0 , \tag{A1}$$

where $\rho(x)=xe^{-x/2}$. We will first solve this problem in the Hilbert space $\mathcal{F}_\rho$ of all square summable functions $\psi(x)$ on the positive half-axis which are orthogonal to ρ. Then we will use the result to get the solution on the space of summable functions. We use Dirac's bracket notation for the inner product in $\mathcal{F}_\rho$, i.e.,

$$\langle\psi|\varphi\rangle=\int_0^\infty \bar{\psi}(x)\varphi(x)dx .$$

Using spectral decomposition, the time dependent problem (A1) reduces to the eigenvalue problem

$$-\frac{\partial^2\varphi_k}{\partial x^2}=k^2\varphi_k \tag{A2}$$

in $\mathcal{F}_\rho$. The eigenfunctions are given by

$$\varphi_k(x)=\left[\frac{2}{\pi}\right]^{1/2}\sin(kx-\theta_k) , \tag{A3}$$

where θ_k is determined by the constraint

$$\langle\rho|\varphi_k\rangle=0 .$$

This equation is easily solved; it yields

$$e^{-2i\theta_k}=\frac{\hat{\rho}(k)}{\hat{\rho}(-k)} ,$$

where

$$\hat{\rho}(k)=\int_0^\infty \rho(x)e^{-ikx}dx=\frac{1}{(1/2+ik)^2}$$

is the Fourier transform of $\rho(x)$. Since the eigenvalue problem (A2) is not self-adjoint, its eigenfunctions are not orthogonal. A simple calculation shows that in fact

$$\langle\varphi_k|\varphi_{k'}\rangle=\delta(k-k')+g(k)g(k') , \tag{A4}$$

where

$$g(k)=\left[\frac{2}{\pi}\right]^{1/2}\frac{k}{1/4+k^2} ,$$

satisfies the normalization condition

$$\int_0^\infty |g(k)|^2 dk=1 . \tag{A5}$$

A similar calculation gives the completeness relation

$$\int_0^\infty dk\,|\varphi_k\rangle\langle\varphi_k|=I+|f\rangle\langle f| , \tag{A6}$$

with

$$f(x)=\sqrt{2}\left[\frac{x}{2}-1\right]e^{-x/2} , \quad \langle f|f\rangle=1 .$$

Inverting the right hand side of Eq. (A6) gives the formula

$$I=\int_0^\infty dk|\varphi_k\rangle\langle\varphi_k|(I-\tfrac{1}{2}|f\rangle\langle f|) ,$$

which allows us to expand an arbitrary initial condition ψ_0 in the eigenbasis. The solution of the initial value problem (A1) is thus given by

$$|\psi_t\rangle=\int_0^\infty dk\, e^{-c^2k^2t}|\varphi_k\rangle \times\langle\varphi_k|(I-\tfrac{1}{2}|f\rangle\langle f|)|\psi_0\rangle . \tag{A7}$$

Let us estimate the norm of this solution. Using the orthogonality relation (A4) we obtain

$$\|\psi_t\|^2=\langle\psi_t|\psi_t\rangle =\int_0^\infty dk\, e^{-2c^2k^2t}|\langle\varphi_k|(I-\tfrac{1}{2}|f\rangle\langle f|)|\psi_0\rangle|^2 +\left|\int_0^\infty dk\, e^{-c^2k^2t}g(k) \times\langle\varphi_k|(I-\tfrac{1}{2}|f\rangle\langle f|)|\psi_0\rangle\right|^2 .$$

Now we apply the Cauchy-Schwarz inequality to the last integral, and make use of (A5) and the fact that

$0 < e^{-c^2k^2t} \le 1$. This gives

$$\|\psi_t\|^2 \le 2\int_0^\infty dk\,|\langle\varphi_k|(I-\tfrac{1}{2}|f\rangle\langle f|)|\psi_0\rangle|^2 .$$

Finally, we use again the completeness formula (A6) to get

$$\|\psi_t\| \le \sqrt{2}\|\psi_0\| . \tag{A8}$$

Thus the solutions of (A1) remain bounded in $\mathcal{F}_\rho$ as $t \to \infty$.

It is also possible to obtain a pointwise estimate of $\psi_t(x)$ for large t. Indeed from (A7), using (A3), we easily get

$$|\psi_t(x)| \le \int_0^\infty dk\,|\langle\varphi_k|(I-\tfrac{1}{2}|f\rangle\langle f|)|\psi_0\rangle| \left[\frac{2}{\pi}\right]^{1/2} \times \int_0^\infty d\xi\, e^{-c^2\xi^2 t} .$$

Using (A3) again we can estimate the inner product in the last formula, and evaluate the integral. This gives

$$|\psi_t(x)| \le \frac{1}{\sqrt{\pi c^2 t}}(1+\tfrac{1}{2}\|f\|_1\|f\|_\infty)\int_0^\infty |\psi_0(y)|dy , \tag{A9}$$

where $\|f\|_1$ and $\|f\|_\infty$ are the L^1 and L^∞ norms of $f(x)$. Thus, for summable initial conditions, the solution $\psi_t(x)$ vanishes uniformly as $t \to \infty$.

The evolution defined by Eq. (A1) for arbitrary function ρ is an interesting question in itself. One can show that as long as $\rho(x)$ decays to zero slower than $x^{3/2}$ at $x=0$ the analysis outlined above holds. We will consider the general case in a future publication.

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