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Dynamical metastability

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Abstract. We present an exact solution for the master equation for a previously studied urn model and use this example as a basis for formulating a general approach to metastability. The emphasis is on dynamics (rather than analyticity) and we also obtain a cutoff partition function that maps smoothly to the usual partition function on passing to the stable state.

The formal problem of metastability in statistical mechanics is due to the competition between two numbers, each of which is ordinarily idealised to be infinite. The large number of particles in macroscopic systems motivates the thermodynamic limit needed for breakdown of analyticity in phase transitions. On the other hand, the long lifetime against homogeneous nucleation in many metastable systems means that there is a reproducible state in which critical droplets do not form on any practical timescale even if the volume is many times the size of a galaxy.

In this paper we return to a previously studied urn model [1] in which dynamic and analytic features were both studied and we describe progress that has now been made on its dynamical aspects, in particular an exact solution for a stationary constant-current state. Various ways of looking at this exact solution are considered, its salient features generalised, and it is suggested that this framework might play a role in the general definition of metastability. We also comment on the relation of our work to a recent discussion of 'bottomless action' potentials [2].

The urn model can be thought of as a model of a cluster or droplet which grows or shrinks one particle at a time. (It can also be thought of as a random walk on the non-negative integers or as a birth and death process.) A cluster of size n is assigned energy E_n and individual particles have chemical potential μ. The Boltzmann factor for an n-particle cluster is thus

$$w_n = \exp(-\beta E_n + \beta\mu n). \tag{1}$$

In [1] the choice $E_n = \log n$ was considered, while for some droplet models $E_n = \sigma n^{(d-1)/d}$ (σ corresponding to surface tension) would be appropriate. Our results in this paper, including the exact solution, do not depend on these details but relate mainly to the convergence properties of sums of w_n and $1/w_n$. We define G_n by

$$w_n = \exp(-G_n) \tag{2}$$

and the partition function by

$$Z = \sum_{n=0}^{\infty} \exp(-G_n). \tag{3}$$

In [1] log Z was observed to be analytic for Re $\mu < 0$. The function was then continued to Re $\mu > 0$ and the relevance to metastability studied. In the present paper analyticity plays only a small role and indeed we believe it would be of interest to remedy that situation.

The dynamics for the urn or cluster are stochastic dynamics. Let X_t be a random variable whose value at time t is the number of balls in the urn or the size of a cluster. The transition probabilities are

$$(P)_{nm} = p_{nm} = \mathrm{prob}(X_{t+1} = m | X_t = n) \tag{4}$$

and are taken to be fixed in time. The matrix P is stochastic, i.e. $\Sigma_m p_{nm} = 1$. The density is

$$(\rho(t))_n = \rho_n(t) = \mathrm{prob}(X_t = n) \tag{5}$$

and satisfies the master equation

$$\rho_n(t+1) = \sum_m \rho_m(t) p_{mn} \qquad \text{or} \qquad \rho(t+1) = \rho(t)P. \tag{6}$$

The transition probabilities are assumed to satisfy detailed balance

$$w_n p_{nm} = w_m p_{mn} \tag{7}$$

which is enough to guarantee that $\rho_n = w_n/Z$ is a formal invariant solution of (6). The dynamics therefore give the correct equilibrium distribution when equilibrium is possible. For the P we study, if Z is not finite, 1 is no longer in the spectrum of P and the largest eigenvalue will then be smaller than 1. (In some cases, infinite Z may still allow 1 to be in the continuum spectrum of P, but we do not consider this possibility for the moment.)

There is considerable leeway in the choice of $\{p_{nm}\}$ such that it be consistent with (7). We use

$$\begin{aligned} p_{nm} &= \alpha(w_m/w_n)^{1/2} = \alpha \exp[\tfrac{1}{2}(G_n - G_m)] && |n-m| = 1 \\ p_{nm} &= 0 && |n-m| > 1 \\ p_{00} &= 1 - p_{01} && \\ p_{nn} &= 1 - \sum_{k \neq n} p_{nk} && n \geqslant 1. \end{aligned} \tag{8}$$

For appropriate $\{w_n\}$ there exists positive α so that $p_{nn} > 0$ for all n ($\{|G_{n+1} - G_n|\}$ should be uniformly bounded). The square root of w will make frequent appearances and we define $u_n = (w_n)^{1/2}$. We further define $U = \mathrm{diag}(u_0, u_1, \ldots)$ and

$$B = \frac{1}{\alpha}[1 - UPU^{-1}] = \begin{pmatrix} b_0 & -1 & & & 0 \\ -1 & b_1 & -1 & & \\ & -1 & \cdot & & \\ & & & \cdot & \\ 0 & & & & \cdot \end{pmatrix} \tag{9}$$

where $b_0 = (w_1/w_0)^{1/2}$, $b_n = (w_{n+1}/w_n)^{1/2} + (w_{n-1}/w_n)^{1/2}$, $n \geqslant 1$. $(u_0, u_1, \ldots)$ is a formal eigenvector of B with eigenvalue 0 and corresponds to the formal invariant left eigenvector w_n/Z of P. Now, however, the criterion for being in the point spectrum is *square* integrability. Generally for the Hermitian B the natural environment is L_2, rather than L_1, as it is for P. We consider situations where w_n diverges sufficiently rapidly that 0 is not in the spectrum of B at all, and we seek the lowest eigenvalue, designated $\beta_g=(1-e^{-\gamma})/\alpha$. In [1] a bound of order $\mu^{\beta+1}$ (as $\mu\to 0$) was found and numerical evidence presented for the bound being attained. The importance of this eigenvalue is that it governs the long time exponential decay of the system. This can be seen from the spectral decomposition of B and by recalling the definition of B (9) in terms of P.

In this paper we shift the question slightly so as to find an exact answer for the eigenvalue and eigenfunction. We also use the exact answer to get a perturbation theory value of β_g.

Consider

$$f_n = u_n S_n$$

with

$$S_n=\sum_{j=n}^{\infty}(w_j w_{j+1})^{-1/2}. \tag{10}$$

For $n\geq 1$ we have

$$\begin{aligned}(Bf)_n &= -f_{n-1}+[(w_{n+1}/w_n)^{1/2}+(w_{n-1}/w_n)^{1/2}]f_n-f_{n+1}\\ &=0.\end{aligned} \tag{11}$$

However

$$(Bf)_0=(w_1/w_0)^{1/2}f_0-f_1=(w_0)^{-1/2}=1. \tag{12}$$

There are two ways to rephrase the question so that (10) becomes its exact answer. Let δB be the matrix whose only non-zero element is $(\delta B)_{00}=-1/S_0$. Then as before, for $n\geq 1$, $((B+\delta B)f)_n=0$. Now however

$$((B+\delta B)f)_0=1+(\delta B)_{00}f_0=0.$$

Therefore f is an exact eigenvector of $B+\delta B$ with eigenvalue 0. An estimate of β_g follows by treating $-\delta B$ as a perturbation of $B+\delta B$ so that

$$\beta_g\sim\frac{\langle f|(-\delta B)|f\rangle}{\langle f|f\rangle}=\frac{f_0^2/S_0}{\Sigma f_n^2}=\frac{1}{S_0\,\Sigma\, w_n(S_n/S_0)^2}. \tag{13}$$

Two interesting quantities make their appearance in (13). Define

$$Z_c=\sum_{n=0}^{\infty} w_n S_n^2/S_0^2. \tag{14}$$

This will later be seen to serve as a cutoff partition function in the metastable regime and for the situations considered in [1] (and for $\beta\geq 2$) is continuous at $\mu=0$. On the other hand, S_0 will generally be divergent at the transition, and for the urn model

$$S_0=\sum_{j=0}^{\infty}(w_j w_{j+1})^{-1/2}\sim\int_0^{\infty}\mathrm{d}x\, x^{\beta}\exp(-\mu\beta x)=(\beta\mu)^{-(\beta+1)}\Gamma(\beta+1). \tag{15}$$

This gives β_g a μ dependence of $\mu^{\beta+1}$ (for $\beta>2$), in agreement with [1].

Another and perhaps more fruitful way to look at f is as a way to get a stationary solution to

$$\rho(t+1)=\rho(t)P+Q \tag{16}$$

with Q a source term. From our calculations we know that $(Uf)^\dagger$ will be a time-independent solution of (16) if Q possesses a zero component only (thus $Q = (q_0, 0, \ldots)$), but its magnitude will depend on appropriate normalisation of f. Let $f' = Cf$ with C a normalisation constant. C will be fixed by requiring that Uf' go over to w_n/Z in passing from the metastable to the stable state. We shall later argue for Z_c being the appropriate replacement for Z. Assuming this to be the case, the relation

$$w_n/Z_c = (Uf')_n = Cw_nS_0$$

implies

$$C = \left(S_0 \sum w_n (S_n/S_0)^2 \right)^{-1}. \tag{17}$$

With this C, the vector $\xi = Uf'$ is a suitably normalised stationary solution of (16). Substituting, we have

$$Q = \xi^\dagger P - \xi^\dagger = C\alpha(1, 0, 0, \ldots).$$

ξ is therefore a steady state probability density, appropriately normalised for small n, for a case where there is a steady release of particles at the origin. This is an alternative view of metastability in which one does not watch for decay (nor try to define what has decayed and what has not) but rather asks at what rate must new systems be introduced to keep the overall distribution constant. It is an approach that has been taken in previous studies of metastability in which dynamics are emphasised; see, for example, the discussion of Landauer and Swenson's and Langer's work in [3, ch 6]. With this interpretation the quantity $C\alpha$ is identified with the decay rate γ and using (17) we have

$$\gamma = C\alpha = \alpha(S_0 Z_c)^{-1} \tag{18}$$

with Z_c given by (14). This coincides with the perturbation results (13).

One can turn things around and take (16) as the defining equation for both stable and unstable regimes. The usual situation is recovered in the stable regime precisely because γ vanishes (or $S_0 \to \infty$) as one goes to the stable side. Thus when $\Sigma\, w_n$ exists, S_0 does not exist and we have an invariant state. On the other hand, when S_0 exists, $\Sigma\, w_n$ does not and the constant current solution is obtained. Whether or not the quantity $\Sigma_{n=0}^{\infty}\, \xi_n$ exists depends on whether or not the escape time to infinity is finite. In the ϕ^4 potential of [2] that time is finite: in the urn model of [1] it is not. Moreover, even when $\Sigma\, \xi_n < \infty$, its moments, such as $\Sigma\, n^2 \xi_n$, may be infinite. This would seem to present a problem of interpretation but in fact it is not $\{\xi_n\}$ that will be suggested as the candidate for the metastable state. (As usual, 'state' means the probability distribution over configurations.)

In looking at the magnetisation of what was termed the metastable state in a stochastic dynamics for the Ising model, McCraw and Schulman [4] took systems that had not 'decayed' at some fixed T and averaged the magnetisation over the interval $[T/2, T]$. (Systems that had decayed were used to compute lifetimes.) A system was considered to have decayed when its magnetisation fell below a predetermined value, macroscopically distant from both the stable value and the value about which it hovered prior to decay. (Zero magnetisation could be taken here to avoid the impression of circularity.) In effect, the definition of the metastable state in [4] involved not only the probability that the system had a given configuration at some t, but also a conditioning that required that at a later time T it had not decayed.

For the definition that we propose we use a sharper version, suggested by Aizenman and Newman [5], of the above conditioning. Let $\theta(n;\, N, T)$ be the probability that the urn has n balls at T given that it had 0 balls at 0 and that at no time in the interval 0 to $2T$ did it contain more than N balls (note the 2 in the condition). Next let $T\to\infty$ and finally $N\to\infty$. The resulting θ_n is proposed as the metastable state. Thus

$$\begin{aligned}\theta(n;\, N, T) &= \text{prob}(X_T = n | X_t \leq N \text{ for } 0 \leq t \leq 2T \text{ and } X_0 = 0)\\ &= \frac{\text{prob}(X_T = n \text{ and } X_t \leq N \text{ for } 0 \leq t \leq 2T | X_0 = 0)}{\text{prob}(X_t \leq N \text{ for } 0 \leq t \leq 2T | X_0 = 0)}\end{aligned}$$

where we have rewritten the conditional probability. Now break the interval in half and use the fact that once the urn has n balls (at T) the fact that $X_t \leq N$ for $t<T$ is irrelevant for $t>T$ by the Markov property. Therefore

$$\theta(n;\, N, T) = \frac{\begin{array}{c}\text{prob}(X_T = n \text{ and } X_t \leq N \text{ for } 0 \leq t \leq T | X_0 = 0)\\ \times \sum_{j=0}^{N} \text{prob}(X_{2T} = j \text{ and } X_t \leq N \text{ for } T \leq t \leq 2T | X_T = n)\end{array}}{\sum_{i=0}^{N} \text{prob}(X_{2T} = i \text{ and } X_t \leq N \text{ for } 0 \leq t < 2T | X_0 = 0)}. \tag{19}$$

Each term in each product in the numerator of (19) represents a T step transition probability (from 0 to n and from n to j) under a modified dynamics in which exit from $[0, N]$ is not allowed. Transition probabilities for a single step under this dynamics are given by P^N, the truncation of P to its first $N+1$ rows and columns. The denominator represents the same evolution for a period $2T$. Therefore

$$\theta(n;\, N, T) = \sum_{j=0}^{N} ((P^N)^T)_{0n}((P^N)^T)_{nj}\left(\sum_{i=0}^{N} ((P^N)^{2T})_{0i}\right)^{-1}. \tag{20}$$

The matrix P^N is not stochastic and its largest eigenvalue is less than 1. Call this largest eigenvalue $\exp(-\gamma_0^N)$ and note that it is related to the lowest eigenvalue of the corresponding truncated B (called B^N and equal to $U(1-P^N)U^{-1}/\alpha$) by

$$\exp(-\gamma_0^N) = 1 - \alpha\beta_0^N. \tag{21}$$

The eigenvalues and eigenvectors of B^N are denoted $\beta_k^N = [1-\exp(-\gamma_k^N)]/\alpha$ and f^{kN}, respectively. Using the spectral decomposition of B^N, (20) becomes

$$\theta(n;\, N, T) = \frac{\sum_{k,j=0}^{N} \exp[-T(\gamma_k^N + \gamma_j^N)] f_0^{kN} f_n^{jN*} \sum_{i=0}^{N} f_i^{jN*} u_i}{\sum_{k=0}^{N} \exp(-2T\gamma_k^N) f_0^{kN} \sum_{i=0}^{N} f_i^{kN*} u_i}. \tag{22}$$

For $T\to\infty$ and appropriate $\{w_n\}$ the $k=j=0$ term in (22) dominates. Thus

$$\theta(n;\, N, \infty) = f_n^{0N*} f_n^{0N}. \tag{23}$$

If we further assume appropriate spectral properties for B ($=\lim_N B^N$) we obtain

$$\begin{aligned}\theta_n &= \lim_{N\to\infty} \lim_{T\to\infty} \theta(n;\, N, T)\\ &= f_n^{0*} f_n^0\end{aligned} \tag{24}$$

with f^0 the ground-state eigenvector of B. This is basically the same as the probability distribution proposed in [2].

f^0 is not equal to our previously defined f, nor is γ_0 ($=\lim_{N\to\infty}\gamma_0^N$) the same as γ, but they are close. Formally this is easy to see. As noted above f satisfies

$$(Bf)_n = \delta_{0n}. \tag{25}$$

Since 0 is not in the B spectrum, we invert (25) to obtain

$$f=\sum_{k=0}^{\infty} \beta_k^{-1} f^k f_0^{k*}. \tag{26}$$

If we assume that close to the transition $\beta_0 \to 0$ and that the sum in (26) is dominated by the $k=0$ term we find

$$f \simeq \beta_0^{-1} f^0 f_0^{0*}. \tag{27}$$

It follows that f and f^0 are proportional and taking into account the normalisation of f^0 we have

$$f^0=f(\sum |f_n|^2)^{-1/2}. \tag{28}$$

This yields

$$\theta_n=\frac{w_n S_n^2}{\Sigma\, w_k S_k^2}=\frac{w_n (S_n/S_0)^2}{\Sigma\, w_k (S_k/S_0)^2}. \tag{29}$$

Based on (29) we view $w_n(S_n/S_0)^2$ as a modified Boltzmann factor and the denominator

$$Z_c=\sum w_n (S_n/S_0)^2$$

which provides normalisation, as a partition function. For the urn model S_n/S_0 remains close to unity for $n \ll 1/\mu$ and for $n \gg 1/\mu$ drops off like $1/w_n$, as we will now show, using the method of steepest descents on the integral (15) with appropriately modified limits of integration.

In particular, for the urn model we approximate S_n by

$$I(x)=\int_x^{+\infty} \exp(\beta \log \xi - \beta\mu\xi)\, \mathrm{d}\xi \qquad \text{for } \mu>0.$$

The maximum of the argument of the exponent is at $\xi_0 = 1/\mu$ and its first and second derivatives are, respectively, $\beta(1/\xi - \mu)$ and $-\beta/\xi^2$. We then obtain the asymptotic expressions

$$I(x) \sim \frac{\exp(-\beta \log \mu - \beta)}{(\beta\mu^2)^{1/2}} \qquad \text{for } x \ll 1/\mu$$

$$I(x) \sim \frac{\exp(\beta \log n - \beta\mu n)}{\beta(1/x - \mu)} \qquad \text{for } x \gg 1/\mu.$$

(Note that for small x, $I(x)$ is nearly independent of x.) It follows that

$$f_n = u_n S_n \sim u_n I(x) \sim C u_n \qquad \text{for } n = x \ll 1/\mu$$

and

$$f_n \sim C'/w_n \qquad \text{for } n = x \gg 1/\mu.$$

The state $\theta_n \sim f_n^2$ is thus indistinguishable from the pseudo-equilibrium state w_n for $n \ll 1/\mu$ (μ small) and for $n \gg 1/\mu$, it decreases like $(w_n)^{-1}$. This is another indication of the suitability of θ_n as a metastable state. The near constancy of S_n, followed by its rapid dropoff, will generally be true of droplet models.

θ_n is thus our candidate for the metastable state and the fact that $S_n/S_0 \simeq 1$ for n less than the droplet size $(=1/\mu)$ reflects the general prejudice that the metastable state ought to resemble a stable state if a droplet has not formed. What has always been difficult in attempts to make this idea precise has been the problem of where to draw the line. Various criteria for the acceptance of some system configurations and the exclusion of others have been proposed [6] but have generally involved elements of arbitrariness and discontinuity as a function of external field (corresponding to μ in the urn problem). The factor $(S_n/S_0)^2$, on the other hand, provides diminishing significance to states that violate the droplet condition but in a smooth fashion and in a way that automatically accommodates increasing droplet size as the phase transition is approached from the metastable side.

Several remarks are in order on the possibility that this example may have broader significance.

(i) At this stage the role of analyticity is not evident to us. In (18) we have an expression for γ, the decay rate, but do not see a simple relation of this to the analytic continuation of the free energy, an expression for which was given in [1] (based on [7]). Supposedly γ is related to Im(free energy).

(ii) The two-time conditioning does not seem adequate to pin down the metastable state in the Ising model [5]; apparently one's intuition about what is likely and unlikely is not always a guide to the infinite volume limit. This suggests that the best possible results for metastability may prove to be the asymptotics as one approaches the transition. Hints of this already occur in considerations [8, especially § 5] of the putative formula $\gamma = \kappa \operatorname{Im} f$. However, the enormous volumes that would be needed before the aforementioned intuitions are violated suggest that the domain of applicability of the asymptotic expressions should be large, at least away from critical points.

(iii) Theoretical description of spinodal decomposition is problematic and approaches in which singularities of the free energy are sought deep in the metastable region (inspired perhaps by the van der Waals gas [see 9, 10]) have not proved useful, probably representing too great an extrapolation from the stable regime.

With emphasis on the dynamics one looks to properties of P or B and here we wish to suggest that in some instances the onset of spinodal decomposition corresponds to the disappearance of the point spectrum at the bottom of the spectrum of B. The decay would no longer be dominated by a single exponential and the ground-state eigenvector of B, which gave a state with time-independent properties up until its decay, would no longer exist. It is likely that this has something to do with properties of critical droplets but our present conjecture has the advantage of not relying on a specific picture of nucleation or decay.

(iv) Although we anticipate problems in going to the infinite degree of freedom Ising or field theoretical case, the generalisation of our results from one to n dimensions, $1 < n < \infty$, is straightforward. In the continuum case, for example, one looks to the Fokker–Planck equation with a source

$$\partial\rho/\partial t = \operatorname{div}(\operatorname{grad}\rho + \rho \operatorname{grad} G) + Q. \tag{30}$$

Q here is the rate γ times a delta function and one would set $\operatorname{grad}\rho + \rho\operatorname{grad} G$ equal to a divergence-free vector in order to obtain the constant current solution. Alternatively, the transformation $f = \rho\exp(G/2)$ would yield a Schrödinger-like equation for f.

A solution to the one-dimensional version (the Schrödinger analogue) of (30) that corresponds to f_n of (10) is

$$f(x) = \exp(-G(x)/2)\int_x^\infty \mathrm{d}y\,\exp(G(y)). \tag{31}$$

Solutions of this sort have generally been rejected as stationary states [11, p 98] because the corresponding density $\rho = f\exp(-G/2)$ is not normalised. (Practically speaking, however, the solution (31) is used both in [3] and [11] for the calculation of decay rates, much as we have done.) The suggestion considered in this paper and in [2] is that it is $f(x)^2$ that should be considered the metastable probability distribution, and this is normalisable. The n-dimensional generalisation of this idea is obvious, although, as indicated above, $n=\infty$ may introduce new problems.

(v) The dynamical rule (8) is not the only one for which an exact solution is available. For example, one can also satisfy detailed balance with the choice $p_{n\,n-1} = \alpha w_{n-1}/w_n$, $p_{n\,n+1} = \alpha$, with other definitions in (8) unchanged. For the steady state solution one replaces S_n (of 10) by the even simpler $\Sigma_{j=n}^{\infty}(w_j)^{-1}$. These solutions may be of interest in specific problems and in multidimensional situations one's guesswork may be aided by considering more easily found continuum solutions (of (30)).

(vi) We reconsider the case $\mu<0$. We know that w_n/Z is the normalised stable state and u_n a square summable eigenstate of B of eigenvalue 0. We want to construct a Green function for B with a source at 0 and a sink at a particular point n. Thus we seek a vector v_j with

$$(Bv)_j = \delta_{0,j} - \tau\delta_{n,j} \tag{32}$$

where τ is some positive constant. Define

$$\sigma_k = \sum_{j=0}^{k-1}\left(\frac{1}{w_j w_{j+1}}\right)^{1/2} \qquad k\geqslant 1$$

and $\sigma_0 = 0$. Then to satisfy (32), including continuity at n, we must have

$$v_k = u_k(\tau\sigma_n u_n - \sigma_k) + c' u_k \qquad k\leqslant n$$

$$v_k = u_k(\tau u_n - 1)\sigma_k + c' u_k \qquad k\geqslant n$$

where c' is any constant. The condition that this state is square summable is that $\tau = 1/u_n$. In that case, we take c' so that v_k is orthogonal to the fundamental state u_k, namely

$$c'Z = \sum_{k=0}^{n-1} w_k(\sigma_k - \sigma_n).$$

In summary, the Green function with source at 0 and sink at n orthogonal to the fundamental state satisfies

$$(Bv)_j = \delta_{0,j} - \left(\frac{1}{u_n}\right)\delta_{n,j}$$

$$v_k = u_k\frac{\Sigma_{j=0}^{n-1} w_j(\sigma_j - \sigma_n)}{Z} - (\sigma_k - \sigma_n) \qquad k\leqslant n$$

$$v_k = u_k\frac{\Sigma_{j=0}^{n-1} w_j(\sigma_j - \sigma_n)}{Z} \qquad k>n.$$

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