Reaction rates and the noisy saddle-node bifurcation: Renormalization group for barrier crossing

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Barrier crossing calculations in reaction-rate theory typically assume a large barrier limit. When the barrier vanishes, however, there is a qualitative change in behavior. Instead of crossing a barrier, particles slide down a sloping potential. We formulate a renormalization group description of this transition and derive the universal scaling behavior and corrections to scaling for the escape time in overdamped systems with arbitrary barrier height. Our critical theory unifies barrier crossing in chemistry with the renormalization group, and with bifurcation theory for discrete chaotic maps.

In this letter, we investigate deep connections between barrier crossing, the renormalization group, and the noisy saddle node bifurcation. In particular, we show that Kramers’ reaction rates can be understood as an asymptotic limit of the universal scaling near the continuous transition between high-barrier and barrier-less regimes. Applying methods from stochastic processes theory we derive an analytical expression for the universal scaling function for the mean barrier escape time near the critical point, giving the crossover between high and low barrier limits. The renormalization group provides a framework within which this result can be understood and systematically improved by perturbative calculations of corrections to scaling, some of which we give explicitly.

Barrier crossing arises in applications across physics, chemistry, and biology. In 1940, Kramers computed the barrier crossing rate for particles in both overdamped and underdamped regimes [1]. This result and others [2–4] provided the theoretical explanation for the Arrhenius equation describing chemical rate coefficients

\[ k \approx \exp\left(-\frac{E_b}{k_B T}\right) \]

where \( E_b \) is the energy barrier for activation [5]. More recent efforts have established the escape rate at arbitrary damping, giving the crossover between the low- and high-damping limits [6, 7], and have accounted for the effects of state-dependent [8, 9], non-gaussian [10–12], and colored [12–14] noise, anharmonic corrections [15, 16], and fluctuating barriers [14, 17].

Most transition-state calculations assume a large barrier limit. This means the barrier escape is a rare event, with a separation of time scales between the escape and relaxation into a quasi-equilibrium state before crossing [18]. In the limit of vanishing barrier, however, there is a qualitative change in behavior. Particles instead slide down a monotonic potential, spending the most time near its inflection point. To capture the low barrier escape rate, extensions to Kramers’ theory have been developed, incorporating anharmonic corrections for instance, but these have significant errors when the barrier and thermal energy are comparable (\( E_b \approx k_B T \)) [15].

Finite barrier escape problems have garnered increasing theoretical interest over the past decade, with several studies contributing further low barrier refinements of existing theories [19–24] or focusing directly on the saddle-node bifurcation where the barrier vanishes [25, 26]. Such escape processes are relevant to certain high precision measurements. For instance, force spectroscopy experiments apply a force on a single bond in a biomolecule until it breaks [20, 27]. For typical molecules, the critical force, at which the energy barrier for breaking vanishes and Kramers’ theory breaks down, is now well within the reach of atomic force microscopy and optical tweezers [27]. Another exciting application is in micro- and nano-electromechanical devices, which sensitively switch oscillation amplitude in response to an input signal by operating near the barrier-less critical point [25, 28]. Here, an analytical theory of low barrier crossing would help to distinguish between noise and signal activated switching.

We develop a critical theory for barrier crossing with a renormalization group approach that gives a complete scaling description of the noisy saddle-node bifurcation. We are inspired by previous work on the intermittency [29] route to chaos [30–32], where the renormalization group coarse-grains in time, then rescales the system to fix a certain term in the potential. In chaos theory, this procedure involves iterating and rescaling a discrete map [31, 32], leading to a different fixed point for the same renormalization group equations used by Feigenbaum to study period doubling [33]. We take the continuous time limit, reducing the renormalization group to a series of elementary rescalings and yielding a simplified description applicable to barrier escape problems. Our procedure organizes what amounts to dimensional analysis, providing an elegant framework that unifies Kramers’ theory for Arrhenius barrier crossing with the renormalization group and the noisy saddle-node bifurcation.

As a starting point, we consider the equation of motion for a overdamped particle in a general potential \( V(x) \) and driven by spatially dependent white noise,

\[ \dot{x} = f(x) + g(x) \xi(t). \]  \hspace{1cm} (1)

Here \( f(x) = -\eta^{-1} dV/dx \) is the force exerted on the particle (divided by the damping coefficient \( \eta \)) and \( g(x) \) is the spatially varying noise amplitude (with the damping absorbed). The noise \( \xi(t) \) has zero mean, \( \langle \xi(t) \rangle = 0 \) and
is uncorrelated in time, $\langle \xi(t)\xi(t') \rangle = \delta(t-t')$. With barrier crossing phenomena in mind, we consider potentials with boundary conditions $V(x) \to \infty$ as $x \to -\infty$ and $V(x) \to -\infty$ as $x \to \infty$. The potential either has a single barrier or is monotonically decreasing (e.g. Figure 1). The quantity of interest is the mean barrier crossing time $\tau$, defined as the time particles take to reach $+\infty$ from an initial position at $-\infty$.

Besides the experimental systems discussed above, this model also serves as the natural description for a general chemical reaction, involving the transition between metastable species $A$ and $B$. These species are points in a $3N$ dimensional configuration space defined by the locations of $N$ reaction constituents. Coarse-graining to a one dimensional reaction coordinate, which parameterizes the minimal gradient path between the states $A$ and $B$, neglecting effects of memory friction and noise correlations, and taking the overdamped limit produces Eq. (1). The effective potential along the reaction coordinate has a barrier separating species $A$ and $B$ (a detailed derivation is given by Hänggi et al. [18]).

We parameterize Eq. (1) by the Taylor coefficients of $g(x)$ and $f(x)$,

$$\frac{dx}{dt} = \sum_{n=0}^{\infty} c_n x^n + \xi(t) \sum_{n=0}^{\infty} g_n x^n.$$  

(2)

The renormalization group defines a flow in this space of systems described by a single reaction coordinate. Near the renormalization group fixed point, the behavior is most effectively described by a single Taylor expansion at the origin. In contrast, for large barriers in Kramers’ theory, the escape time is characterized by two expansions, capturing the harmonic oscillations in the potential well and at the top of the barrier. These two equivalent schemes are shown in Fig. 1. Given the later expansion at the two extrema, the expansion at the origin parameterizing perturbations away from the fixed point is uncorrelated in time, $\langle \xi(t)\xi(t') \rangle = \delta(t-t')$. With barrier crossing phenomena in mind, we consider potentials with boundary conditions $V(x) \to \infty$ as $x \to -\infty$ and $V(x) \to -\infty$ as $x \to \infty$. The potential either has a single barrier or is monotonically decreasing (e.g. Figure 1). The quantity of interest is the mean barrier crossing time $\tau$, defined as the time particles take to reach $+\infty$ from an initial position at $-\infty$.

![FIG. 1. Typical potentials in the high barrier Arrhenius limit (solid curve) and at the renormalization group fixed point (dashed curve). Kramers’ theory utilizes a two point series expansion at $x_{min}$ in the potential well and at $x_{max}$, the top of the barrier. For our renormalization group approach the natural description is in terms of a single expansion at the origin parameterizing perturbations away from the fixed point potential $V^\ast(x) \propto -x^3$. Also shown is the noise amplitude $g(x)$, which generically has spatial dependence (dotted curve).](image)

For a generic analytic potential this happens when the barrier vanishes and $V(x) = -x^3$ is locally a perfect cubic. Therefore, we rescale our system to fix the coefficient $c_2$, corresponding to the cubic term in the potential. The correct rescaling defines a new spatial coordinate $\tilde{x} = bx$. After both coarse graining and rescaling, we arrive at

$$\frac{d\tilde{x}}{dt} = \sum_{n=0}^{\infty} c_n b^{2-n} \tilde{x}^n + \tilde{\xi}(t) \sum_{n=0}^{\infty} g_n \tilde{x}^n.$$  

(3)

We can then read off how the parameters flow under the renormalization group, $\tilde{c}_n = b^{2-n} c_n$ and $\tilde{g}_n = b^{3/2-n} g_n$. These flows and exponents exactly match those found under the discrete-time renormalization group [31, 32], indicating that the scaling of the 'intermittency route to chaos' [29] is also non-anomalous [36]. Taking the coarse graining factor to be close to 1, $b = (1 + d\ell)$, we obtain continuous flow equations,

$$\frac{dc_n}{d\ell} = (2-n) c_n, \quad \frac{dg_n}{d\ell} = (3/2-n) g_n.$$  

(4)

The eigenfunctions of the renormalization group in our continuum theory are the monomials $x^n$ and noisy monomials $\xi(t)x^n$. If the right hand side of Eq. (2) is an eigenfunction, it is scaled by a constant factor under the action of the renormalization group. These eigenfunctions
are the much simpler continuous time limit of those for
the discrete-time renormalization group [32]. In partic-
ular, the cubic potential \( V(x) \propto -x^3 \) (without noise)
is the fixed point. At the fixed point, particle trajec-
tories \( x(t) \sim 1/t \) exhibit scale invariance in time as they
approach the cubic inflection point at \( x = 0 \). Perturba-
tions away from the fixed point lead to dynamics with
non-power law decay to a locally stable state or over the
inflection point.

The mean barrier crossing time is a function of the po-
tential shape and the noise correlation, encoded through
the expansion coefficients \( \epsilon_n \) and \( g_n \). Thus, the problem asymptotically reduces to finding the
contribution to the linear term in the expansion of \( ˜V \)
be modeled as a cubic potential with a linear perturba-
tion away from the fixed point lead to dynamics with

\[
\tau = g_0^{-2/3} T \left( \{ \epsilon_n/g_0^{2(n-3)/3} \}, \{ g_n/g_0^{1-2n/3} \} \right),
\]

where \( T \) is a universal function of a single variable \( T \).

When the scaling form Eq. (5) could have been written
down using dimensional analysis, the renormalization group
approach provides the natural structure and moti-
vation for our approach. The parameter space flows
indicate that, with a fixed quadratic force, the constant
and linear force and noise terms \( \{ \epsilon_0, \epsilon_1, g_0, g_1 \} \) are rel-
vant, growing under coarse graining and dominant on
long time scales. Other variables are irrelevant and can
be incorporated perturbatively. Of the relevant variables,
the linear force coefficient \( \epsilon_1 \) can be set to zero by plac-
ing the origin at the inflection point of the potential.

The spatial dependence of the noise (including the rel-
vant linear term \( g_1 \)) can also be removed by a change of co-
ordinates \( x \to \tilde{x} \) with \( \tilde{x} \) defined by [37]

\[
x = \int_{\tilde{x}}^{\tilde{\tilde{x}}} \frac{g_0}{g(y)} dy,
\]
producing a system with constant noise \( \tilde{g}(\tilde{x}) = g_0 \) and
force \( \tilde{f}(\tilde{x}) = f(\tilde{x})/g(\tilde{x}) \) (hence \( g_1 \) was relevant because it con-
tributed to the linear term in the expansion of \( \tilde{f} \)).

Systems near enough to the critical point therefore can be
modeled as a cubic potential with a linear perturbation
\( V(x) = -x^3/3 - \epsilon_0 x \) and constant noise \( g_0 \). The
escape time scaling form becomes,

\[
\tau = g_0^{-2/3} T(\epsilon_0/g_0^{4/3}).
\]

Thus, the problem asymptotically reduces to finding the
universal function of a single variable \( T(\alpha) \), where \( \alpha = \epsilon_0/g_0^{4/3} \). The limiting form of the scaling function \( T(\alpha) \)
must give the known solutions. In the limit \( \alpha \to -\infty \) the
barrier is large compared to the noise, so the Kramers
approximation applies and we have that [1],

\[
T(\alpha) \sim \frac{\pi}{|\alpha|^{1/2}} e^{\frac{1}{2}|\alpha|^{3/2}}.
\]

For our choice of parameters, the energy barrier is given
by \( E_b/k_B T = 8/3|\alpha|^{3/2} \). In the opposite limit \( \alpha \to \infty \),
the potential is downward sloping with gradient much
larger than the noise level. The passage of particles over
the inflection point occurs even in the absence of noise
(in contrast to the Kramers limit, which requires noise for
barrier escape). Therefore, the crossing time approaches
that for a deterministic particle in the cubic potential.

One can easily show that the limiting scaling form is

\[
T(\alpha) \sim \frac{\pi}{\alpha^{1/2}}.
\]

We now turn our focus to obtaining an exact analytical
expression for \( T(\alpha) \) that is valid for all \( \alpha \).

To this end, we study the trajectories of particles in-
jected at position \( x_i \) and time \( t_i \) into a general potential
\( V(x) \) with noise \( g_0 \) and compute the mean first passage
time to \( x_f \), following the standard approach [18, 30, 38].

Let \( P(x,t) \) be the distribution of particles over positions
\( x \) at time \( t \), with \( P(x,t) = \delta(x - x_f) \). The probability
that a particle has not reached \( x_f \) at time \( t \) is

\[
P(t) = \int_{-\infty}^{x_f} P(x,t) dx.
\]

Note that \( P(0) = 1 \) and \( P(t) \to 0 \) as \( t \to \infty \) as long
as there is noise driving the system, which guarantees
particles reach \( x_f \). The distribution of first passage times
is \( p(t) = -dP/\partial t \) so that the mean first passage time is

\[
\tau(x_i) = \int_0^\infty p(t) dt = \int_0^\infty P(t) dt,
\]

where we integrate by parts for the second equality. To
derive a differential equation for \( \tau(x_i) \), we start from the
Kolmogorov backward equation for distribution \( P(x,t) \) with
initial condition \( x_i [39] \),

\[
-\frac{dP(x,t)}{dt_i} = -V'(x_i) \frac{dP(x,t)}{dx_i} + \frac{1}{2} g_0^2 \frac{d^2 P(x,t)}{dx_i^2}.
\]

To write this equation in terms of the mean first passage
time \( \tau \), we multiply both sides by \( t \) and integrate over \( x \) and \( t \). Using the relations in Eqs. (10) and (11) and the
identity \( dP(x,t)/dt_i = -dP(x,t)/dt \), we arrive at

\[
\frac{1}{2} g_0^2 \tau''(x_i) = V'(x_i) \tau'(x_i) = -1.
\]

This gives a simple ordinary differential equation for the
first passage time from \( x_i \) to \( x_f \) of particles in potential
\( V(x) \) and constant noise with amplitude \( g_0 \). The bound-
ary conditions are \( \tau(x_f) = 0 \) and \( \tau'(\infty) = 0 \), which
encode absorbing and reflecting boundaries respectively.

Writing the solution to Eq. (13) in integral form, we ar-
rive at the result obtained in Refs. [18, 30, 38],

\[
\tau(x_i) = \frac{2}{g_0} \int_{x_i}^{x_f} dy \int_{-\infty}^{y} dz \exp \left[ \frac{\alpha}{g_0^2} (V(y) - V(z)) \right],
\]
which satisfies the boundary conditions as long as \( V'(x) \to -\infty \) as \( x \to -\infty \). For large barriers, it is known that Eq. (14) reproduces Kramers escape rate formula via a saddle point approximation that expands the potential around the maximum and the minimum (as shown in Fig. 1) to second order [18].

Our renormalization group analysis allows us to restrict our focus to the relevant variables. For the cubic potential (systems on the unstable manifold of the renormalization group fixed point), the escape time can be computed analytically using Eq. (14) in the limit \( x_f = -x_i \to \infty \). We find that \( \tau = g_0^{-2/3} T(\alpha) \) with the universal scaling function given by

\[
T(\alpha) = 2^{1/3} \pi^2 \left[ \text{Ai}^2(-2^{2/3} \alpha) + \text{Bi}^2(-2^{2/3} \alpha) \right],
\]

where \( \text{Ai}(x) \) and \( \text{Bi}(x) \) are the first and second Airy functions and \( \alpha = \epsilon_0/g_0^{4/3} \) as above. This solution is shown in Figure 2, along with the Arrhenius and deterministic limits given in Eqs. (8) and (9) respectively and the mean barrier crossing times from direct simulations of the Langevin process [Eq. (1)]. The universal scaling function \( T(\alpha) \) reproduces the two known limits when the barrier is large or the potential is strongly downward sloping and agrees excellently with the numerical results.

Kramers’ escape rate for the cubic potential follows from Eq. (15) and the asymptotic form of the second Airy function. As \( \alpha \to 0 \), however, contributions from the first Airy function become important so that Kramers’ theory and extensions involving anharmonic corrections break down. The difference between Eqs. (8) and (15) is also related to the narrowing of the spectral gap of the barrier crossing Fokker-Plank operator (which has been measured numerically [40]).

The scaling function Eq. (15) also serves as a starting point from which the theory can be systematically improved by computing corrections to scaling. The higher order terms in the potential are irrelevant variables under the renormalization group flows and hence can be treated perturbatively. For instance, consider a quartic perturbation \( \delta V(x) = -\epsilon_3 x^4/4 \) and let \( \beta = \epsilon_3 g_0^{2/3} \). In the Kramers regime, \( \alpha \to -\infty \), we have that \( T(\alpha, \beta) \approx T(\alpha) + \beta^2 T_3(\alpha) \) to leading order, where

\[
T_3(\alpha) \xrightarrow{\alpha \to 0} \pi \sqrt{|\alpha|} \epsilon_3^2 |\alpha|^{3/2} (8|\alpha|^{3/2} + 11)/8. \quad (16)
\]

In the deterministic regime, \( \alpha \to \infty \), we also add a quintic term as a regulator on the boundary conditions of the potential, \( \delta V(x) = -\epsilon_4 x^5/5 + \epsilon_4 x^5/5 \), with \( \epsilon_4 > 0 \) and sufficiently large so that the potential remains monotonically decreasing. To quadratic order in \( \beta \) and \( \gamma = \epsilon_4 g_0^{2/3} \), the universal scaling function is [41]

\[
T(\alpha, \beta, \gamma) \xrightarrow{\gamma \to 0} T(\alpha) + \beta^2 \left( \frac{15}{8} \pi \sqrt{|\alpha|} - \frac{3\pi}{4\sqrt{\gamma}} \right) - \pi \sqrt{\gamma} + \frac{3}{2} \pi \sqrt{\alpha} \gamma - \frac{5}{2} \pi \alpha \gamma^{3/2} + \frac{35}{8} \pi \alpha^{3/2} \gamma^2.
\]

The term \( \pi/\sqrt{\alpha} \) is just the deterministic limit of the scaling form for the cubic potential and \( \beta^2 T_3(\alpha) = -15\pi \sqrt{|\alpha|} \beta^2/8 \) comes from the quartic perturbation to the inflection point. Other terms arise from quintic corrections or global changes in the potential. Here \( \gamma \) is a dangerous irrelevant variable [42, Sections 3.6, 5.4, & 5.6], which has a pole \( 3\pi\sqrt{\beta^2}/4\sqrt{\gamma} \) in the expansion about 0, because it is needed to keep the potential monotonic (for \( \beta \neq 0 \)).

We expect our results will be directly applicable to barrier crossing processes in which thermal fluctuations are comparable to the energy barrier including the aforementioned experimental systems, narrow escape problems in cellular biology [43], and downhill protein folding scenarios [44, 45]. A more thorough analysis of incorporating perturbative corrections from irrelevant variables into Eq. (14) would be both theoretically interesting and useful in applications. It would also be useful to study the applicability of our renormalization group analysis to systems with colored noise, multiple dimensions, or in other damping regimes. The effects of colored noise are encoded in the correlation function \( \langle \xi(t) \xi(t') \rangle = G(x, t - t') \). The renormalization group transformation can be adapted to act on the Fourier transform of this quantity \( \hat{G}(x, \omega) \), giving flows of the colored noise under coarse-graining. For some reactions, an underdamped
model or multi-dimensional reaction coordinate may be required for an accurate description. Renormalization group scaling will provide a natural organizing framework for these studies.

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