Breaking a chain of particles: the role of mass and inter-particle potential

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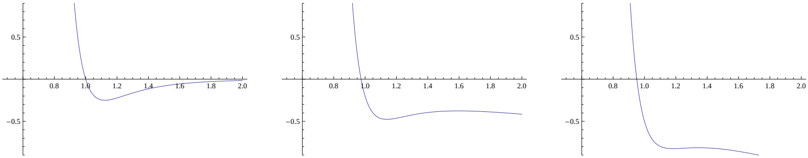
A problem from dynamic force spectroscopy

- Idea: stretch a molecular bond until it breaks, measure the force needed.
- Gives information about bond strength.
- Noise enters via thermal fluctuations, measurement errors etc.
- Model: ($U =$ potential with minimum a, $r =$ loading rate):

$$\mathrm{d}y_s = \Big(- U'(y_s) + rs \Big)\mathrm{d}s + \sigma \mathrm{d}W_s, \qquad y_0 = a$$

- Exit problem from a time dependent domain.



Adiabatic approximation: Kramer's rate theory

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Assume U has a local maximum at b.

For time-independent potential, and small noise, the rate of escape from a potential barrier of height $E_0 = U(b) - U(a)$ is

$$k = A_0 \exp\Big(-\frac{E_0}{2\sigma^2} \Big).$$

Exponential rate: [Arrhenius 1889]. Prefactor: [Eyring 1935, Kramers 1940].

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Exponential rate: [Arrhenius 1889]. Prefactor: [Eyring 1935, Kramers 1940].

Now assume that the loading rate is also small, and use the instantaneous rate at all times. Put (with $F = rt$)

$$P(t) = \mathbb{P}(\text{bond survived until time t}),$$
$$P(F) = \mathbb{P}(\text{bond survived until force reaches F}).$$

Then

$$\frac{\mathrm{d}}{\mathrm{d}t} P(t) = -k(t)P(t), \qquad \frac{\mathrm{d}}{\mathrm{d}F} P(F) = -\frac{1}{r} k(F) P(F).$$

Adiabatic approximation: rate constant

$$\frac{\mathrm{d}}{\mathrm{d}F}P(F) = -\frac{1}{r}k(F)P(F), \qquad H(y,F) = U(y) - Fy$$

Then,

$$P(F) = \exp\Big(-\frac{1}{r}\int_0^F k(F')\mathrm{d}F'\Big).$$

Question: how to approximate the time-dependent rate $k(F')$?

Recall:

$$k(F) \sim \mathrm{e}^{-E(F)/2\sigma^2}, \qquad E(F) = H_{\max}(F) - H_{\min}(F).$$

[Bell '78] First order expansion around F-independent min and max:

$$H(y,F) \approx U(y_\pm) + \frac{1}{2}U''(y_\pm)(y-y_\pm)^2 - Fy, \quad E(F) \approx E_0 + F\Delta.$$

[Garg '95] Second order expansion around inflection point: $F_\mathrm{c} = U'(y_\mathrm{c})$,

$$H(y,F) \approx \big(U(y_\mathrm{c}) - F_\mathrm{c}y_\mathrm{c}\big) - (F - F_\mathrm{c})q + \tfrac{1}{6}U'''(y_\mathrm{c})(y-y_\mathrm{c})^3$$

$$E(F) \approx \mathrm{const}(1 - F/F_\mathrm{c})^{3/2}$$

[Lin et. al., PRL 98 (2007)], [Fridddle, PRL 100 (2008)].

Adiabatic approximation: consclusions

- Adiabatic approximation means use of large deviation estimates.
- The Garg model is a mixture of LD and small energy barrier assumption.
- The details of the potentials do not matter, only certain characterstics do.
- All models are overdamped.

Surely, the honest way to treat the problem is to investigate a SDE and consider the distribution of the first exit time from a domain. This has not been done as far as we know. We do it for a different model and a different question...

The basic model

Consider a chain of three particles. One is fixed at $x = 0$, and one is pulled at speed ε. Their ideal position is at mutual distance a.

$\boldsymbol{x}(s) = (0, x_s, 2a(1 + \varepsilon s)) \in \mathbb{R}^3$ are the positions of the particles.

The middle particle satisfies

$$\mathrm{d}x_s = -\frac{\partial H}{\partial x}(x_s, \varepsilon s)\mathrm{d}s + \sigma \mathrm{d}W_s$$

with initial condition $x_0 = a$ and time-dependent potential energy given by

$$H(x, \varepsilon s) = U(x) + U(2a(1 + \varepsilon s) - x).$$

U is a pair potential.

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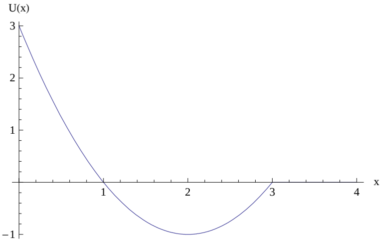
Question 1: On which side does the chain break first? Note that the answer is obvious in the deterministic case.
Question 2: To what extent does the potential (modelling assumption) matter?

Potentials and breaking criteria

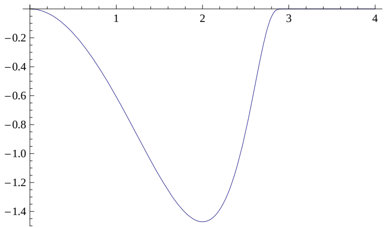
First possibility: Convex, compact support, e.g.:

$$U(y) = \begin{cases} (|y| - a)^2 - (b-a)^2 & 0 \leqslant y \leqslant b \\ 0 & \text{otherwise} \end{cases}$$



Second possibility: Smooth, compact support, e.g.:

$$U(y) = \begin{cases} -y^2 \, \mathrm{e}^{-1/(3-y)} & 0 \leqslant y \leqslant 3 \\ 0 & \text{otherwise} \end{cases}$$



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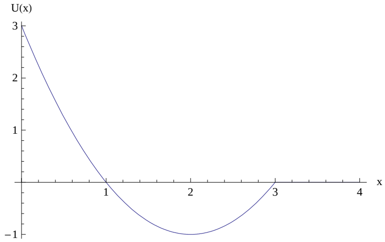

Break when particle hits support boundary ($\Rightarrow$ exit time)

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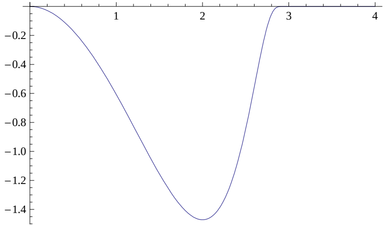

Break determined by the long time behaviour of the particle.

Potentials and breaking criteria

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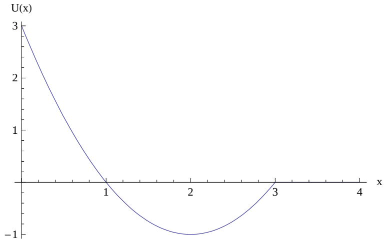

Break when particle hits support boundary
($\Rightarrow$ exit time)

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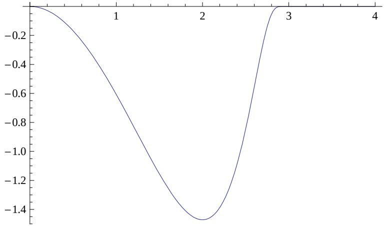

Break determined by the long time behaviour of the particle.

Important difference: In second case, the particle is almost free just before the chain breaks!

Convex potentials: results

$$\mathrm{d}x_s = -\frac{\partial H}{\partial x}(x_s, \varepsilon s)\mathrm{d}s + \sigma \mathrm{d}W_s$$

Stopping time $\tau = \inf\{t \geqslant 0 : x_t \notin (2a + t - b, b)\}$.
Break condition sets:

$$L = \{x \in C_a([0,\infty)) : x_\tau = b\}, \qquad R = C_a([0,\infty)) \setminus L.$$

Theorem (Allmann, B. '09)

1. *(Fast pulling):*
$$\sigma\sqrt{|\ln\sigma|} \ll \varepsilon \ll 1 \quad \Longrightarrow \quad \lim_{\sigma\to 0} \mathbb{P}(R) = 1.$$
2. *(Slow pulling):*
$$\exp(-\sigma^{-2/3}) \ll \varepsilon \ll \sigma\sqrt{|\ln\sigma|}^{-1} \quad \Longrightarrow \quad \lim_{\sigma\to 0} \mathbb{P}(R) = 1/2.$$

Note: the threshold is (roughly) $\varepsilon = \sigma$.

Smooth potentials: Breaking criteria

$$\mathrm{d}x_s = -\frac{\partial H}{\partial x}(x_s, \varepsilon s)\mathrm{d}s + \sigma \mathrm{d}W_s$$

Problem: How to characterize breaking?
Idea: Look at long time behaviour. But in the original model this does not make sense. Our solution: Local version of the evolution (relative to the midpoint).

$$\mathrm{d}x_s = -(x_s^3 - \varepsilon s x_s - \varepsilon)\mathrm{d}s + \sigma \mathrm{d}W_s$$

Start the equation at $s = -\infty$. Put

$$L = \{x : \lim_{t\to\infty} x_t = -\infty\}, \quad R = \{x : \lim_{t\to\infty} x_t = \infty\}.$$

Smooth potentials: Results

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Theorem (Allmann, B., Hairer '10)

1. *(Fast pulling):*

   $$\sigma^{4/3}|\ln\sigma|^{2/3} \ll \varepsilon \ll 1 \quad \Longrightarrow \quad \lim_{\sigma\to 0} \mathbb{P}(R) = 1.$$

2. *(Slow pulling):*

   $$\sigma^2|\ln\sigma|^3 \ll \varepsilon \ll \sigma^{4/3}\sqrt{|\ln\sigma|}^{-13/6}$$

   $$\Longrightarrow \quad \lim_{\sigma\to 0} \mathbb{P}(R) = \lim_{\sigma\to 0} \mathbb{P}(L) = 1/2.$$

Conclusion: more noise needed to randomize the break location when pulling at speed ε.

Convex case: Rescaling, centering, localizing

$$\mathrm{d}x_s = -\frac{\partial H}{\partial x}(x_s, \varepsilon s)\mathrm{d}s + \sigma \mathrm{d}W_s$$

Rescale time $t = \varepsilon s$ to get

$$\mathrm{d}x_t = -\frac{1}{\varepsilon}(U'(x_t) + U'(2a + t - q_t))\mathrm{d}t + \frac{\sigma}{\sqrt{\varepsilon}}\mathrm{d}W_s$$

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Now center around the deterministic solution: $y_t = x_t - x_t^{\mathrm{det}}$, and write

$$\mathrm{d}y_t = \frac{1}{\varepsilon}(A(t)y_t + b(y_t, t))\mathrm{d}t + \frac{\sigma}{\sqrt{\varepsilon}}W_t,$$

with

$$A(t) = -U''(x_t^{\mathrm{det}}) - U''(2a + t - x_t^{\mathrm{det}}), \qquad |b(y,t)| \leqslant My^2.$$

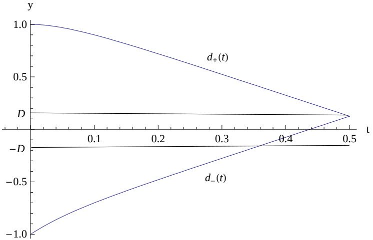
$-A(t)$ is bounded above and away from zero, and so we can compare with an Ornstein-Uhlenbeck process!

Convex case: Proof idea

The exit boundary forms a space-time triangle, where the tip is offset from zero by order ε. The variance of the process y_t is of order

$$\frac{\sigma^2}{\varepsilon}\mathbb{E}\Big(\int_0^t \mathrm{e}^{(s-t)/\varepsilon}\,\mathrm{d}W_s\Big)^2 = \frac{\sigma^2}{\varepsilon}\int_0^t \mathrm{e}^{2(s-t)/\varepsilon}\,\mathrm{d}s \approx \sigma^2.$$

This gives the threshold $\sigma = \varepsilon$.



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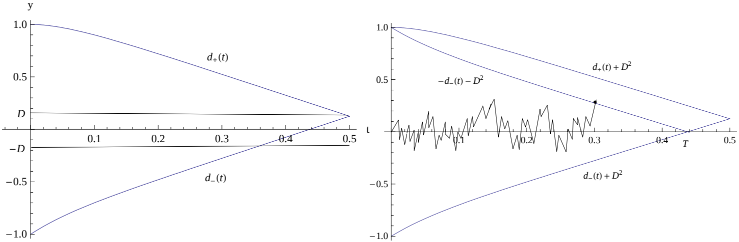

The actual proof is more involved and uses techniques by Nils Berglund and Barbara Gentz.

Smooth case: deterministic solution versus diffusion

$$\mathrm{d}x_t = -\Big(\frac{1}{\varepsilon}(x_t^3 - tx_t) - 1\Big)\mathrm{d}s + \frac{\sigma}{\sqrt{\varepsilon}}\mathrm{d}W_t$$

For the deterministic solution x_t^{det}, we have

$$x_t^{\mathrm{det}} \asymp \begin{cases} \varepsilon/|t| & \text{for } t \leqslant -\sqrt{\varepsilon} \\ \sqrt{\varepsilon} & \text{for } -\sqrt{\varepsilon} \leqslant t \leqslant \sqrt{\varepsilon} \end{cases}$$

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The process is approximately free Brownian motion during $-\sqrt{\varepsilon} \ll t \ll \sqrt{\varepsilon}$, and very constrained before that.
So, at $t = \sqrt{\varepsilon}$, we have

$$\mathbb{E}(x^2_{\sqrt{\varepsilon}}) \approx \frac{\sigma^2}{\varepsilon}\mathbb{E}(W^2_{\varepsilon^{1/2}}) = \frac{\sigma^2}{\varepsilon^{1/2}}$$

So the standard deviation is $\sigma\varepsilon^{-1/4}$. For this to be greater than $\sqrt{\varepsilon}$ we need $\sigma \gg \varepsilon^{3/4}$.

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So the standard deviation is $\sigma\varepsilon^{-1/4}$. For this to be greater than $\sqrt{\varepsilon}$ we need $\sigma \gg \varepsilon^{3/4}$.
The actual proof is unfortunately much more involved, and heavily uses (and modifies) the machinery of Berglund and Gentz.

Massive particles: model

$$\begin{aligned} \mathrm{d}q_t &= p_t \, \mathrm{d}t \\ \varepsilon^{\beta} \mathrm{d}p_t &= -p_t \, \mathrm{d}t + \frac{1}{\varepsilon}(t \, q_t - q_t^3 + \varepsilon) \, \mathrm{d}t + \varepsilon^{\alpha} \, \mathrm{d}W_t \end{aligned}$$

- Describes the deviation from the midpoint of the massive particle, approximates the potential to fourth order.
- The mass of the particle is ε^{β}.
- When compared to the previous diffusion constant $\sigma\varepsilon^{-1/2}$, we have assumed the form $\sigma = \varepsilon^{\alpha+1/2}$.
- Initial conditions are such that $\lim_{t\to-\infty} q_t = \lim_{t\to-\infty} p_t = 0$.

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- Initial conditions are such that $\lim_{t\to-\infty} q_t = \lim_{t\to-\infty} p_t = 0$.
- Question: Does the mass influence the break probability?
- Recall: for $\beta = \infty$ we have a change of behaviour at $\alpha = 1/4$.

Massive particles: small mass

$$\mathrm{d}q_t = p_t\,\mathrm{d}t$$
$$\varepsilon^{\beta}\mathrm{d}p_t = -p_t\,\mathrm{d}t + \frac{1}{\varepsilon}(t\,q_t - q_t^3 + \varepsilon)\,\mathrm{d}t + \varepsilon^{\alpha}\,\mathrm{d}W_t$$

Theorem (ABH '10)

Assume $\beta > 2$. *There exist* $c_1, \gamma > 0$ *such that for* $t_1 = c_1\sqrt{\varepsilon|\ln\sigma|}$ *and any* $t_2 > t_1$,

1. *(Fast Pulling) if* $\alpha > 1/4$ *then*

$$\lim_{\varepsilon\to 0}\liminf_{s\to-\infty}\mathbb{P}^s\left\{\inf_{t_1\leqslant t\leqslant t_2}\frac{q_t}{\sqrt{t}} > \gamma\right\} = 1\,,$$

2. *(Slow Pulling) if* $0 < \alpha < 1/4$ *then*

$$\lim_{\varepsilon\to 0}\limsup_{s\to-\infty}\mathbb{P}^s\left\{\inf_{t_1\leqslant t\leqslant t_2}\frac{q_t}{\sqrt{t}} > \pm\gamma\right\} = 1/2$$

Small mass: proof idea

$$\begin{aligned} \mathrm{d}q_t &= p_t \, \mathrm{d}t \\ \varepsilon^\beta \mathrm{d}p_t &= -p_t \, \mathrm{d}t + \frac{1}{\varepsilon}(t\, q_t - q_t^3 + \varepsilon) \, \mathrm{d}t + \varepsilon^\alpha \, \mathrm{d}W_t \end{aligned}$$

The idea is a comparison with two overdamped dynamics: For a set of paths approaching measure one as $\varepsilon \to 0$, we show

$$q_t^- \leqslant q_t + \varepsilon^\beta P_t \leqslant q_t^+$$

with

$$\mathrm{d}q_t^\pm = \frac{1}{\varepsilon}(t q_t^\pm - (q_t^\pm)^3 + \varepsilon(1 \pm \mathcal{O}(\varepsilon))) \, \mathrm{d}t + \varepsilon^\alpha \, \mathrm{d}W_t$$

with W_t the same Brownian motion that drives the massive equation, and

$$P_t = \varepsilon^{\alpha-\beta} \int_{-T}^{t} \mathrm{e}^{-(t-s)\varepsilon^{-\beta}} \, \mathrm{d}W_s$$

Small mass: proof idea

$$\mathrm{d}q_t = p_t\,\mathrm{d}t$$
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But what about larger mass?

Large mass: a linear model

$$\begin{aligned} \mathrm{d}q_t &= p_t \,\mathrm{d}t \\ \varepsilon^{\beta}\mathrm{d}p_t &= -p_t \,\mathrm{d}t + \frac{1}{\varepsilon}(t\, q_t - q_t^3 + \varepsilon)\,\mathrm{d}t + \varepsilon^{\alpha}\,\mathrm{d}W_t \end{aligned}$$

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- Note the drastically changed behaviour for $t \to \infty$.
- The breaking condition is now (intuitively) trivial.

Large mass: a linear model

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- Note the drastically changed behaviour for $t \to \infty$.
- The breaking condition is now (intuitively) trivial.
- There is an explicit solution:

$$\begin{aligned} q(t) =& \pi \varepsilon^{(1-2\beta)/3} \Big(- \mathrm{Ai}(t(\varepsilon, \beta)) \int_{-\infty}^{t} \mathrm{e}^{-\frac{1}{2}(t-s)\varepsilon^{-\beta}} \, \mathrm{Bi}(s(\varepsilon, \beta))(\mathrm{d}s + \varepsilon^{\alpha} \mathrm{d}W_s) \\ & + \mathrm{Bi}(t(\varepsilon, \beta)) \int_{-\infty}^{t} \mathrm{e}^{-\frac{1}{2}(t-s)\varepsilon^{-\beta}} \, \mathrm{Ai}(s(\varepsilon, \beta))(\mathrm{d}s + \varepsilon^{\alpha} \mathrm{d}W_s) \Big) \end{aligned}$$

with $s(\varepsilon, \beta) = \varepsilon^{-(1+\beta)/3}(s + \varepsilon^{1-\beta}/4)$.

Large mass: a linear model

$$\mathrm{d}q_t = p_t\,\mathrm{d}t$$
$$\varepsilon^\beta \mathrm{d}p_t = -p_t\,\mathrm{d}t + \frac{1}{\varepsilon}(t\,q_t \qquad + \varepsilon)\,\mathrm{d}t + \varepsilon^\alpha\,\mathrm{d}W_t$$

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$$+ \operatorname{Bi}(t(\varepsilon,\beta))\int_{-\infty}^{t} \mathrm{e}^{-\frac{1}{2}(t-s)\varepsilon^{-\beta}}\operatorname{Ai}(s(\varepsilon,\beta))(\mathrm{d}s + \varepsilon^\alpha \mathrm{d}W_s)\Big)$$

with $s(\varepsilon,\beta) = \varepsilon^{-(1+\beta)/3}(s + \varepsilon^{1-\beta}/4)$.

- The second term is asymptotically dominant.

Linear model: convergence for large times

$$q(t) = \pi\varepsilon^{(1-2\beta)/3}\Big(-\mathrm{Ai}(t(\varepsilon,\beta))\int_{-\infty}^{t} \mathrm{e}^{-\frac{1}{2}(t-s)\varepsilon^{-\beta}}\,\mathrm{Bi}(s(\varepsilon,\beta))(\mathrm{d}s+\varepsilon^{\alpha}\mathrm{d}W_s)$$

$$+\,\mathrm{Bi}(t(\varepsilon,\beta))\int_{-\infty}^{t} \mathrm{e}^{-\frac{1}{2}(t-s)\varepsilon^{-\beta}}\,\mathrm{Ai}(s(\varepsilon,\beta))(\mathrm{d}s+\varepsilon^{\alpha}\mathrm{d}W_s)\Big)$$

Lemma (ABH '10)

Put

$$\tilde{q}(t) = \frac{1}{\pi\varepsilon^{(1-2\beta)/3}}\frac{\mathrm{e}^{\frac{1}{2}t\varepsilon^{-\beta}}}{\mathrm{Bi}(t(\varepsilon,\beta))}q(t)\,.$$

Then $\lim_{t\to\infty}\tilde{q}(t)$ *exists almost surely, and is a Gaussian random variable with mean* $m = \varepsilon^{(1+\beta)/3}\,\mathrm{e}^{-\frac{1}{12}\varepsilon^{1-2\beta}}$ *and variance*

$$v = \varepsilon^{2\alpha+(1+\beta)/3}\,\mathrm{e}^{-\frac{1}{4}\varepsilon^{1-2\beta}}\int_{-\infty}^{\infty}\mathrm{e}^{s\varepsilon^{(1-2\beta)/3}}\,\mathrm{Ai}(s)^2\,\mathrm{d}s\,.$$

Asymptotics of Airy integrals

$$m = \varepsilon^{(1+\beta)/3}\,\mathrm{e}^{-\frac{1}{12}\varepsilon^{1-2\beta}}$$

$$v = \varepsilon^{2\alpha+(1+\beta)/3}\,\mathrm{e}^{-\frac{1}{4}\varepsilon^{1-2\beta}} \int_{-\infty}^{\infty} \mathrm{e}^{s\varepsilon^{(1-2\beta)/3}}\,\mathrm{Ai}(s)^2\,\mathrm{d}s\ .$$

Put

$$J(p) = \int_{-\infty}^{\infty} \mathrm{e}^{2ps}\,\mathrm{Ai}^2(s)\,\mathrm{d}s\ .$$

Lemma

There exist constants c_1 *and* c_2 *such that*

(i) $\lim_{p\to\infty} p^{1/2}\,\mathrm{e}^{-2p^3/3}\,J(p) = c_1$,

(ii) $\lim_{p\to 0} p^{1/2}\,\mathrm{e}^{-2p^3/3}\,J(p) = c_2$.

Asymptotics of Airy integrals

$$m = \varepsilon^{(1+\beta)/3}\, \mathrm{e}^{-\frac{1}{12}\varepsilon^{1-2\beta}}$$

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So, $m(\varepsilon)/\sqrt{v(\varepsilon)} = \mathrm{const}\ \varepsilon^{-\alpha+1/4}$, independent of β!

Linear model: result and discussion

$$\mathrm{d}q_t = p_t\,\mathrm{d}t$$
$$\varepsilon^\beta \mathrm{d}p_t = -p_t\,\mathrm{d}t + \frac{1}{\varepsilon}(t\,q_t + \varepsilon)\,\mathrm{d}t + \varepsilon^\alpha\,\mathrm{d}W_t$$

Theorem (ABH '10)

If $\alpha > 1/4$ *then* $\lim_{\varepsilon\to 0}\liminf_{s\to-\infty}\mathbb{P}^s\{\lim_{t\to\infty} q_t = +\infty\} = 1$.

If $\alpha < 1/4$ *then* $\lim_{\varepsilon\to 0}\liminf_{s\to\infty}\mathbb{P}^s\{\lim_{t\to\infty} q_t = \pm\infty\} = 1/2$.

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- This seems to be too strong to be expected in general.
- For example, when starting at finite negative time with zero initial condition, β starts to play a role: the threshold is $\alpha = (1 + \min\{\beta, 0\})/4$, works up to $\beta = -1$.
- Not clear how much of this survives the addition of a fourth order term in the potential.