Supporting Information

The purpose of this material is to put the random walk on parabolas process described in section 2 of the main paper on rigorous mathematical footing. It has been relegated here so that the physical concepts discussed in the paper are not obscured by mathematical details. Throughout the following, the stochastic process $\{A_q\}_{q\in\mathbb{R}_+}$ is denoted by A, and A_t is the value of A at time t. Assume a complete filtered probability space $(\Omega, \mathscr{F}, \mathscr{F}_t, P)$ and that the horizontal and vertical coordinates of the incoming atom are $\mathscr{F}_t$-adapted, continuous path stochastic processes, which are denoted as X and Y, respectively. Furthermore, we assume that the paths of X and Y are sufficiently smooth for the velocity processes $\{dX_t/dt\}$ and $\{dY_t/dt\}$ to exist, are of finite variation on compacts, and that the initial conditions X_0 and Y_0 are given. All of these are obvious from a physical point-of-view. For mathematical convenience, we suppose that the atom moves in the bounded hemisphere $H = \{(x, y) \in \mathbb{R}^2 : \sqrt{x^2 + y^2} \leq \sqrt{X_0^2 + Y_0^2}\}$.

Basic definitions

Definition 1. *Let N_0 be a given countable finite collection of $N(0, \sqrt{k_B T/\pi\gamma})$ and take the subset $B = \{n : n \in N_0, 0 \leq n \leq Y_0\}$. The collection F of all functions from $\mathbb{R}$ to $\mathbb{R}$ with representation*

$$f(x) = b\left(1 - (x - a)^2/h^2\right) \tag{1}$$

where $a \in C$, $C = \left[-\sqrt{X_0^2 + Y_0^2} - h, \sqrt{X_0^2 + Y_0^2} + h\right]$, and $b \in B$ and h a fixed positive real constant, is the local mode collection.

Each of the functions in the local mode collection corresponds to a possible local mode that the incoming particle may pass over during its motion in H. By taking the set N_0 as given, we can be sure that the amplitudes of the local modes (B) are not correlated with the motion of the particle. Countability of this collection ensures its existence. Its finiteness poses no physical limitation as we can make it as large as we wish.

Using $f(x;\alpha,\beta)$ as shorthand for the function $f(x) = \beta\,(1-(x-\alpha)^2/k^2)$ in F, it follows from the definition that

$$C \times B = \{(a,b) : f(x;a,b) \in F \text{ for all fixed } x \in \mathbb{R}\}\,, \tag{2}$$

which means that each element of $C \times B$ corresponds to a unique element of F. The heuristic construction in section 2 of the main paper described a fictional random walker which identified which local mode was directly beneath the incoming atom at time t. By (2), we can assign $C \times B$ as the state space of that walker. Letting Z_t denote its trajectory, we can identify the random walk on parabolas as the process $\{f(X_t;Z_t)\}$.

Definition 2. *The process defined by the random variables*

$$Q_t = f(X_t;Z_t) \tag{3}$$

where $f \in F$ and $Z \mapsto C \times B$ is a pure jump process is a random walk on parabolas.

Each local mode in the local mode collection corresponds to a state of the process Z, and the sojourn periods of Z correspond to the duration that the local mode spends beneath the incoming atom as the atom passes over the surface. This provides an unambiguous connection between the value of Z and a local mode in F, as is more precise than the notation '$f_{Z_t}(X_t)$' used in section 2.

Isotropy and continuity conditions

Before going on to define the process Z, it will be useful to make some comments about the isotropy and continuity conditions. These conditions must be satisfied when Z makes a jump to another region of $C \times B$. In the heuristic construction of section 2, we defined A_{ij}^{+} as the event where $a_j \geq X_t \geq a_i$ when the atom is travelling from left-to-right across the surface, $dX_t/dt > 0$. We required either this condition or A_{ij}^{-} to be fulfilled when the random walk Z made a transition from local mode i to local mode j

at time t. Since the local mode collection F is uncountable in the present construction, it is not so meaningful to speak of 'local mode i' or 'local mode j'. However, we can still describe the situation by considering the value of the C component of the process Z at transition time U. If the value of the C component of Z at time $U- = \lim_{u \to U, u<U} u$ is c_1, then we require the event

$$A_0(c_1) = \left\{ X_{U-} < c_1 \cap \dot{X}_{U-} < 0 \right\} \cup \left\{ X_{U-} > c_1 \cap \dot{X}_{U-} > 0 \right\}, \qquad (4)$$

where $\dot{X}_{U-} = dX_t/dt\,|_{t=U-}$, to occur for a transition to occur at time U. To formulate the second part of the isotropy condition, we suppose that at the transition time U the second component of the process Z takes on the value c_2. Following the construction given in section 2, it must be that the event

$$A_1(c_2) = \left\{ X_U > c_2 \cap \dot{X}_U < 0 \right\} \cup \left\{ X_U < c_2 \cap \dot{X}_U > 0 \right\} \qquad (5)$$

occurs with the transition. Events $A_0(c_1)$ and $A_1(c_2)$ collectively define the isotropy condition, although not in terms of their intersection. For reasons that will become clear in the next section, it is helpful to speak of them separately. The continuity condition is obviously $\{Q_{U-} = Q_U\}$, and so the condition that must occur with probability 1 at the transition time U is

$$A(c_2, b) = A_1(c_2) \cap \{Q_{U-} = Q_U\}, \qquad (6)$$

where b is the value of the amplitude component $b \in B$ of Z after completing the transition.

It is worth pointing out that we can think of the isotropy conditions $A_0(c_1)$ and $A_1(c_2)$ in terms of a symmetry operation on the sample space. To see this, note that events of the form $\{G \cap H^C\} \cup \{H \cap G^C\}$ are usually written as *symmetric differences*,

$$G \triangle H = \{G \cap H^C\} \cup \{H \cap G^C\}.$$

While the events $\{X_{U-} < c_1\}$ and $\{X_{U-} > c_1\}$, and $\{\dot{X}_{U-} < 0\}$ and

$\{\dot{X}_{U-} > 0\}$ are not quite complementary, it is still very tempting to write (4) as a symmetric difference,

$$A_0(c_1) = V_{U-} \triangle C_{U-} \tag{7}$$

where $V_{U-} = \{\dot{X}_{U-} < 0\}$ and $C_{U-} = \{X_{U-} < c_1\}$. Similarly, $A_1(c_2)$ is approximately in the form $\{G^C \cap H^C\} \cup \{G \cap H\}$. By means of de Morgan's laws, it can be shown that such an event can be written as the compliment of a symmetric difference, namely $\{G \triangle H\}^C$. We can therefore rewrite $A_1(c_2)$ as

$$A_1(c_2) = \{V_U \triangle C_U\}^C . \tag{8}$$

We therefore have that $A_1(c_2)$, which occurs at time U, is the complement to events occurring at time $U-$. In this sense, the region of the sample space available at time U is an inversion of the region available at time $U-$. While the association of isotropy with inversion symmetry is indeed very nice, it needs to be regarded with some caution because the representation of $A_0(c_1)$ and $A_1(c_2)$ as symmetric differences and their compliments is not exact.

The pure-jump process Z

The construction will be complete once we define the process Z. This needs to be done in several steps, and the isotropy and continuity condition described above also needs to be considered. It is required that Z makes a jump when either the incoming atom goes on to see a new local mode beneath itself or when the local mode that the atom is currently over begins to take on negative values. We therefore consider two sets of random times $T_1, T_2, \ldots$ and $S_1, S_2, \ldots$, and let $U_i = T_i \wedge S_i$. Now, let $J_1 = T_1, J_2 = T_2 - U_1, \ldots$ be exponentially distributed *iid* random variables, that is

$$P(J_i < j) = 1 - e^{-\lambda j} \tag{9}$$

for some constant $\lambda > 0$. The intervals J_i are the waiting periods between new local modes appearing beneath the incoming atom. The connection

between the parameter λ in (9) and ρ of equation 8 of the main text will be explained shortly. Next, define

$$S_1 = \inf\left(0 < t \leq T_1 : Q_t < 0\right)$$
$$S_2 = \inf\left(U_1 < t \leq T_2 : Q_t < 0\right)$$

and so on. If for a particular S_i such a time does not exist, set it to T_i. The times S_i correspond to times at which the surface profile of the liquid surface begins to take on negative values. For $U_i < t \leq U_{i+1}$, Z is defined as

$$Z_t = \begin{cases} W_{i+1} & t = U_{i+1} \text{ and } A_0(c_1) \text{ occurs} \\ Z_{U_i} & \text{otherwise} \end{cases} \tag{10}$$

where W_{i+1} is a random variable with probability distribution that incorporates the condition $A(c_2, b)$, as will be described shortly. Note that $A_0(c_1)$ will always occur if $U_{i+1} = S_{i+1}$. Thus, the process Z jumps when the particle goes on to see a new local mode beneath itself (an event which occurs after an exponential waiting time and when the isotropy condition is satisfied), or when the local mode begins to take on negative values, whichever occurs first.

To define the probability distribution of the random variable W_{i+1} in (10), recall that there in the random local mode model there is no preference to make any particular transition, as long as the transition satisfies the condition $A(c_2, b)$. We therefore have

$$P(W_{i+1} \in (dc, b)) = \begin{cases} \text{constant} & \text{if } A(c, b) \text{ occurs} \\ 0 & \text{otherwise} \end{cases} \tag{11}$$

where dc is the smallest open interval centered on point c in C.[1] The constant in (11) can, in principle, be evaluated by ensuring that the distribution is normalised over $C \times B$. In this work, we assume that this is the case. Note that the transition probability (11) is defined for a region of C and an individual point in B, rather than for individual local modes like in

[1]The smallest open interval centered on point c is $(c - \epsilon, c + \epsilon)$, $\epsilon \to 0$.

the transition probability in equation 8 of the main text.

From (11) alone, it might not be obvious that the measure $P(W \in dw)$ is mathematically well-defined. This is clarified by the following theorem.

Theorem 1. *The probability measure $P(W \in (dc, b))$ is well-defined.*

Proof. Let $E = \{(c, b) \in C \times B : A(c, b) \text{ is satisfied}\}$. E is the collection of all regions where W may lie with nonzero probability. For $P(W \in (dc, b))$ to be well-defined, it is sufficient for the collection E to correspond to a countable union of disjoint regions in $C \times B$. We can then define $P(W \in E^C)$ as $1 - P(W \in E) = 0$. By the continuity condition in $A((c, b))$, we have

$$f(X_{U_{i+1}}; Z_{U_i}) = f(X_{U_{i+1}}; Z_{U_{i+1}}) = b\left(1 - (X_{U_{i+1}} - c)^2/h^2\right),$$

and hence

$$c = X_{U_{i+1}} - h\sqrt{1 - f(X_{U_{i+1}}; Z_{U_i})/b}, \tag{12}$$

which implies $b \in \left\{\beta : \beta \in B, \beta \geq f(X_{U_{i+1}}; Z_{U_i})\right\}$. By Definition 1, this collection is countable, and by (2) each element corresponds to a unique point $c \in C$. Hence, E corresponds to a countable union of disjoint regions $(dc, b) \in C \times B$.

□

Going further from Theorem 1 and constructing the distribution of Z (and hence of Q) is far from trivial, because the random variables W_i depend upon $X_{U_1}, X_{U_2}, \ldots$. Since we will only be interested in simulating paths of the random walk on parabolas, the above construction is satisfactory.

The final remark is that the parameter of the random walk process Z is the Poisson parameter λ, and that the parameter ρ which featured in the main text is seemingly absent. The latter is used as it is more natural from the point of view of numerical integration of the equations of motion. It is straightforward to show that the pictorial construction closely approximates the proceeding mathematical construction. Suppose that the interval $[U_i, U_{i+1}]$ is partitioned into smaller intervals of length l. If there is a probability ρ during each interval of the walk making a jump, then the probability

that the interval has the length of at least L intervals is $(1-\rho)^L$, and therefore the probability that the jump will occur before time $j = lL$ since the start of the interval is $P(J_{i+1} < j) = 1 - (1-\rho)^L$. Let $L \to \infty$ and $\rho \to 0$, while $L\rho$ is kept at a constant value λj. In this limit

$$\begin{aligned} P(J_{i+1} < j) &= 1 - \lim_{L\to\infty} (1 - \lambda j/L)^L \\ &= 1 - e^{-\lambda j}, \end{aligned}$$

which is the same as (9). Hence, the transition probability in the pictorial construction of section 1 holds well for small time steps and small ρ. As well as its advantage in numerical algorithms, the approximate approach involving ρ will be more lucid than the Poisson parameter in physically-motivated discussions of our work.

Integration with a random walk on parabolas

In order to integrate the equations of motion in the main paper, we need a clear *path-wise* interpretation of the integral $\int_0^t g(Q_t)dQ_t$, where g is a continuous function. In general, special precautions need to be taken when computing integrals with respect to stochastic processes, and it is not usually the case that the results are the same as those of a classical Riemann-Stieltjes integral from ordinary calculus. Such cases arise when the process has paths of *infinite variation*, as well as when the paths are discontinuous. For details, see the reference provided at the end of this section. The paths of the random walk on parabolas process are clearly continuous, so the latter is not an issue. To see that they are of finite variation on compacts, recall that the variation of Q_t on path ω on the interval $[t_a, t_b]$ is

$$V_{[t_a,t_b]}(\omega) = \int_{Q_{t_a}}^{Q_{t_b}} |dQ_t(\omega)| \,. \tag{13}$$

Theorem 2. *The random walk on parabolas process has paths of finite variation on compacts.*

Proof. It will be sufficient to prove the claim on the finite interval $[u_i, u_{i+1}] \subseteq [U_i, U_{i+1}]$, since an arbitrary time interval is covered by a finite union of such intervals. On this interval,

$$Q_t = f(X_t; Z_{U_i})$$

by path continuity, and hence $dQ_t = (df(X_t; Z_{U_i})/dX_t)\, dX_t$. Writing the components of Z_{U_i} as (α, β), (1) gives

$$df(X_t; Z_{U_i})/dX_t = -2\beta(X_t - \alpha)/h^2 \leq 2\beta/h$$

The inequality follows from Definition 2 ($|X_t - \alpha| \leq h$, or else $f < 0$ and $f \notin F$). This gives

$$V_{[U_i,U_{i+1}]}(\omega) = \int_{Q_{U_i}}^{U_{i+1}} |dQ_t(\omega)| \leq (2\beta/h) \int_{X_{U_i}}^{X_{U_{i+1}}} |dX_t(\omega)| \,.$$

Let $\beta \to \max_{b \in B} b$. Since all $b \in B$ are normal random variables, this quantity will be finite for all paths. The finiteness the the right hand side follows from the assumptions that X_t varies smoothly and continuously in C.

□

The crucial consequence of Theorem 2 is that we can integrate the equations of motion with the familiar methods of classical calculus. See the reference below for details.

Reference

Protter, P. E, *Stochastic Integration and Differential Equations*, Springer-Verlag, Berlin, Germany, 2nd edn., 2005.