Simulating sticky particles: A Monte Carlo method to sample a stratification

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January 17, 2020

Abstract

Many problems in materials science and biology involve particles interacting with strong, short-ranged bonds, that can break and form on experimental timescales. Treating such bonds as constraints can significantly speed up sampling their equilibrium distribution, and there are several methods to sample subject to fixed constraints. We introduce a Monte Carlo method to handle the case when constraints can break and form. Abstractly, the method samples a probability distribution on a stratification: a collection of manifolds of different dimensions, where the lower-dimensional manifolds lie on the boundaries of the higher-dimensional manifolds. We show several applications of the method in polymer physics, self-assembly of colloids, and volume calculation in high dimensions.

1 Introduction

Simulating interacting particles is slow: a mere handful of colloidal particles with attractive pairwise interactions can take days or even weeks to collect accurate statistics. The trouble with colloidal particles, and related systems like C₆₀ molecules or atoms with covalent interactions, is they form strong but short-ranged bonds [2,3,17,37,47] causing particles to jiggle rapidly around their current configuration as if they are attached by stiff springs that are gently plucked. Resolving these vibrating springs in a molecular dynamics or Monte Carlo simulation requires tiny time or space steps, steps that are usually much smaller than the scales of the interesting rearrangements, where springs break, form, or change their relative positions.

A natural idea for allowing larger steps is to freeze the vibrations, by adding distance constraints between bonded particles: the springs become rods, which can’t change their lengths. By evolving the system only in directions that preserve the distance constraints, one can take much larger steps, and accelerate the sampling quite significantly. Such ideas are the origin of the popular Shake and Rattle algorithms and their variants in molecular dynamics [2,47], and have led to several Monte-Carlo methods that sample a probability distribution subject to holonomic constraints [7,8,34,35,54,57]. Still, these methods can only be used in limited situations because they require the constraints, or bonds, to be fixed throughout the entire simulation. None allow constraints to be added or dropped, so none can simulate particles with stiff bonds that can break and form.

This paper will introduce a Monte Carlo method to simulate particles with distance constraints that can break and form over the course of the simulation. Its natural application is to study sticky particles, i.e. those that interact only when their surfaces are exactly in contact. A major motivation for developing the method is to simulate DNA-coated colloidal particles (colloids), which are widely studied for their potential use as building blocks for new materials [46]. The sticky approximation works well for DNA-coated colloids [23,44], which can have diameters on the order of 1µm but interact attractively over ranges around 20nm or less, about 2% of the lengthscales of interest [55]. The bonds between DNA-coated colloids must break and form as the system equilibrates, so the vast majority of simulations take small timesteps to resolve the particles’ stiff interactions.

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More generally, the method samples any system with holonomic constraints that can be added or subtracted. Abstractly, it samples a stratification: a union of open manifolds of different dimensions, where the lower-dimensional manifolds lie on the boundaries of higher-dimensional manifolds in a nice enough way [18, 19]. Each manifold in the stratification is assumed to be the level set of a set of scalar functions, the constraints. By adding or removing a constraint, one obtains a manifold with one dimension lower or higher. By forming the union of a collection of manifolds, each defined by a different set of constraints, one obtains a stratification. The method samples a user-defined probability distribution on this stratification.

The configuration space of sticky particles is a specific case of a stratification, if the constraints are the set of pairwise distances between particles. A “manifold” in this stratification is the set of configurations the particles can form subject to a given set of pairwise distance constraints, or bonds. When a bond is added or dropped, this forms a new manifold, whose dimension is one less or more than the previous one. The stratification is the union of all manifolds, or configurations, corresponding to the bonds of interest.

Our method is a natural extension of a well-known sampling algorithm, a random walk Metropolis approximation of the constrained overdamped Langevin dynamics [34, 57]. As in many of the constraint-based methods referenced above, the method sometimes proposes moves that preserve the current set of constraints. However, it additionally attempts to add or subtract constraints: it subtracts each constraint with constant rate, but proposes to add a constraint only when it is close enough to the boundary corresponding to that constraint. For sticky particles, this means that when the surfaces of particles are close together, the method can form a bond between them, and it can always propose to delete a bond. We expect this choice of proposal to be consistent with the dynamics of the system in the limit of small step size, and furthermore, we show that it gives 100% acceptance probability for a stratification consisting of two flat manifolds defined by affine constraints. Overall, the method is a form of reversible jump Monte Carlo (RJMC), a method that is commonly used in Bayesian computations to determine the number of parameters in a model [20]. However, RJMC as it is typically presented requires a parameterization of each of the manifolds that it samples, whereas our method works even without a parameterization.

The method is not the first to try to accelerate simulations of sticky particles. A small number of studies have found clever, but specifically tailored ways to take larger timesteps for particles that are sticky or close to sticky. Seaton & Glandt [48, 49] simulated sticky spheres by choosing one sphere at random and adding or subtracting up to 3 bonds at a time, choosing randomly from all the geometrically possible ways to do so. This method was later improved [31] and used to study the phase diagram of sticky spheres [41]. Another method, “virtual move Monte Carlo,” moves particles with strong but finite-ranged pairwise interactions as a cluster, by displacing one sphere initially, and then moving other spheres if their bond energies changed significantly, and iterating [56]. While these methods are much faster than standard methods, they do not capture the full range of particle rearrangements, and hence, are limited in the kinds of questions they can study. Neither method allows a group of bonded particles to rearrange internally, so neither can adequately study questions that depend on the internal flexibility of a molecule, nor could they approximate the dynamics of particle rearrangement. Furthermore, the method of Seaton & Glandt [48, 49] is tailored specifically to sticky particles, so it is difficult to see how one would adapt it to other kinds of strong, reversible bonds or to more general reversible constraints, since the method requires evaluating difficult integrals that may not have an analytic expression for more general kinds of constraints.

Here is an overview of the paper. Section 2 provides the mathematical setup necessary to describe the sampler: first we describe the specific case of sticky spheres in Section 2.1 and then we give the more general setup for a stratification defined by nearly arbitrary smooth constraints in Section 2.2. We give several examples of stratifications and probability distributions defined on them in Section 2.3. Section 3 presents the algorithm for sampling a probability distribution on a stratification, which we call the Stratification sampler. In Section 4, we revisit the examples defined in Section 2.3 and show a variety of questions one can address using the Stratification sampler. We also show the sampler can be very efficient, sometimes over 1000 times faster than conventional methods, and in addition, we show that it gives a good approximation to a system that is not perfectly sticky. In Section 5 we show our proposal moves give 100% acceptance probability for a stratification consisting of flat manifolds defined by affine constraints with no inequalities. Section 6 discusses some additional applications of the Stratification sampler as well as ideas.
for extending and improving it. Details on how to implement the algorithm are contained in an Appendix.

2 Setup

2.1 Sticky spherical particles

What is the Boltzmann distribution for a system of sticky particles? Consider first $N$ non-sticky particles, thought of as spheres with centers at $x_1, x_2, \ldots, x_N \in \mathbb{R}^d$ and radii $r_1, r_2, \ldots, r_N$. Suppose the energy $U(x)$ of a configuration $x = (x_1, x_2, \ldots, x_N) \in \mathbb{R}^{dN}$ is a sum of pairwise interactions between particles:

$$U(x) = \sum_{i \neq j} U_{ij}(|x_i - x_j|). \quad (2.1)$$

Here $U_{ij}(r)$ is the interaction potential for the pair of particles $(i,j)$, which depends on the distance $r$ between the pair. We could include angular dependence in the potential, but neglect it here for simplicity. The equilibrium probability $\pi(x)$ of a system of particles at temperature $T$ is assumed to be the Boltzmann distribution, $\pi(x) = Z^{-1} e^{-U(x)/k_B T}$, where $k_B$ is Boltzmann's constant. Here $Z$ is the normalization constant, which is finite if the particles are confined in some way. For the sake of presentation, we will assume the particles are restricted to the box $B = [-L, L]^d \subset \mathbb{R}^d$ for $L > 0$, so that $\pi(x) = 0$ for $x \in \mathbb{R}^d - B$.

We are interested in systems for which the attractive parts of the pair potentials are strong and short-ranged. One such a family of potentials is the Morse potential, which depends on parameters $E, \rho, \sigma$ as

$$U_{\text{morse}}(r; E, \rho, \sigma) = E(1 - e^{-\rho(r-\sigma)})^2 - E. \quad (2.2)$$

The Morse potential has an attractive well with minimum depth $E$ at distance $\sigma$ (which for colloids is the sum of the radii), with width proportional to $\rho^{-1}$. For $r < \sigma$, the Morse potential increases steeply, effectively imposing a non-overlap condition between particles. For $r > \sigma + c/\rho$, where $c \approx 2-3$, the potential is nearly zero. The attractive interactions are considered to be strong when $e^E$ is large, and short-ranged when $\rho^{-1} \ll \sigma$.

Another commonly-used model for strongly, short-ranged interactions is the Lennard-Jones($m,2m$) family of potentials, which has a similar structure and similar kinds of parameters [52].

We aim to simulate particles in the sticky limit where the range of the pair potential goes to zero, and the depth goes to infinity, in such a way that the partition function for each bond approaches a constant. Specifically, we require that

$$\int_0^{r_i + r_j + \epsilon} e^{-U_{ij}(r)/k_B T} \, dr \to \kappa_{ij} < \infty \quad \text{as} \quad \epsilon \to 0. \quad (2.3)$$

Here $\epsilon$ is measures the range of the attractive part of the potential, so that $U(r) \approx 0$ for $r \geq r_i + r_j + \epsilon$; for the Morse potential, this occurs around $\epsilon \approx 2.5 \rho^{-1}$ [52]. The constant $\kappa_{ij}$ is the sticky parameter for bond $(i,j)$. It measures the strength of the interaction between particles $(i,j)$: larger $\kappa_{ij}$ means the pair $(i,j)$ spends more time bound when the system is in equilibrium.

We wish to sample the Boltzmann distribution in the sticky limit. To describe the limiting distribution, let $\mathcal{E} = \{(i,j) : i, j = 1, \ldots, N, i \neq j\}$ be the list of all pairs of particles. Define a collection of functions $\Omega = \{q_{ij}(x)\}_{(i,j) \in \mathcal{E}}$ with $q_{ij} : \mathbb{R}^{dN} \to \mathbb{R}$ by

$$q_{ij}(x) = |x_i - x_j|^2 - (r_i + r_j)^2. \quad (2.4)$$

Each function $q_{ij}(x)$ measures the distance between the surfaces of particles $(i,j)$; we call it a bond-distance function. In the sticky limit, the Boltzmann distribution approaches $Z^{-1} e^{-U(x)/k_B T} \to \rho_s(x)$, where the measure $\rho_s(x)$ is defined by [4] [23]

$$\rho_s(x) = Z^{-1} 1_{\mathbb{R}^d}(x) \prod_{(i,j) \in \mathcal{E}} \kappa_{ij} \delta(q_{ij}(x)) + 1, \quad Z = \int_{\mathbb{R}^d} dx \prod_{(i,j) \in \mathcal{E}} \kappa_{ij} \delta(q_{ij}(x)) + 1. \quad (2.5)$$
Here $\delta : \mathbb{R} \to \mathbb{R}$ is a dirac delta-function, and $1_{q \geq 0}(x) = \prod_{(i,j) \in \mathcal{E}} 1_{q_{ij}(x) \geq 0}(x)$ is a product of indicator functions, equal to 0 if there is a pair of particles that is overlapping, and 1 otherwise. This function enforces excluded volume between spheres.

The sticky Boltzmann distribution $\rho_s(x)$ is singular with respect to the Lebesgue measure on $\mathbb{B}$. It concentrates probability on those subsets of configuration space where certain pairs of particles, thought of as spheres, have surfaces that are exactly in contact. To describe these subsets, suppose for the moment that configuration $x$ has no pairs of spheres in contact, so that $x \in M_\emptyset = \{ x \in \mathbb{B} : q_{ij}(x) > 0 \ \forall (i,j) \in \mathcal{E} \}$. The set $M_\emptyset$ is a $dN$-dimensional subset of $\mathbb{B}$, and $\rho_s(x)$ is constant over $M_\emptyset$.

Now suppose that $x$ has one pair of spheres in contact, say pair $(1,2)$. Then $x \in M_{(1,2)}$ where

$$M_{(1,2)} = \{ x \in \mathbb{B} : q_{12}(x) = 0, q_{ij}(x) > 0 \ \forall (i,j) \in \mathcal{E} - \{(1,2)\} \}.$$ 

The set $M_{(1,2)} \subset \mathbb{B}$ is $dN - 1$-dimensional; it has one less intrinsic dimension than the configuration space $\mathbb{B}$. This is because every point in this set is the solution to a scalar equation, so in principle, we could reparameterize $M_{(1,2)}$ using $dN - 1$ variables. (In practice such a reparameterization could be hard to find, and we won’t need to find it.) The measure $\rho_s(x)$ gives finite probability to the set $M_{(1,2)}$—despite it being lower-dimensional than $\mathbb{B}$. Physically, there is a finite probability of finding the system with pair $(1,2)$ exactly in contact.

The same statement holds for all pairs, and we could define sets $M_{(1,3)}, M_{(1,4)}, \ldots, M_{(N-1, N)}$ in a similar way; $\rho_s(x)$ gives finite probability to each of these $dN - 1$-dimensional sets. Moving on, we could also consider the set of configurations where two pairs are in contact, say pairs $(1,2), (2,3)$. This set is given by

$$M_{(1,2),(2,3)} = \{ x \in \mathbb{B} : q_{12}(x) = 0, q_{23} = 0, q_{ij}(x) > 0 \ \forall (i,j) \in \mathcal{E} - \{(1,2), (2,3)\} \}.$$ 

The set $M_{(1,2),(2,3)} \subset \mathbb{B}$ is typically $dN - 2$-dimensional; it has two fewer intrinsic dimensions than $\mathbb{B}$, and in principle could be parameterized by $dN - 2$ variables. The measure $\rho_s(x)$ gives finite probability to the set $M_{(1,2),(2,3)}$, as well as to all other sets where two pairs of spheres are exactly in contact.

In general, given a list $I_{eq} = \{(i_1, j_1), \ldots, (i_m, j_m)\} \subset \mathcal{E}$ of pairs of spheres in contact, and its complement $I_{in} = I_{eq}^\perp = \mathcal{E} - I_{eq}$ of pairs of spheres which satisfy nonoverlap conditions, we define $I = (I_{eq}, I_{in})$ and construct the set

$$M_I = \{ x \in \mathbb{B} : q_{ij}(x) = 0 \ \text{for} \ (i,j) \in I_{eq}, q_{ij}(x) > 0 \ \text{for} \ (i,j) \in I_{in} \}. \quad (2.6)$$

Under certain conditions on the functions in $I_{eq}$ described in the next section, $M_I$ is an open manifold with dimension $dN - m$. The measure $\rho_s(x)$ gives finite, nonzero probability to all manifolds of the form (2.6). Physically, this means that all geometrically possible ways to put particles in contact occur with nonzero probability.

The set formed from the union of all manifolds of the form (2.6) forms a stratification, a geometric object that is more general than a manifold, but still retains some of its nice properties. We may also choose to restrict the number of manifolds in the stratification, by including only a subset of manifolds of the form (2.6). For any given union of manifolds of the form (2.6), our goal is to generate samples from $\rho_s(x)$ restricted to this union.

## 2.2 General mathematical setup

### 2.2.1 Stratification and probability measure

Let’s generalize the system from the previous section, and introduce the notation required to handle more general kinds of constraints. Let $x \in \mathbb{R}^n$, and let $\Omega = \{ q_1(x), q_2(x), \ldots, q_{\mathcal{D}}(x) \}$ be a collection of continuously differentiable scalar functions. For sticky spheres these functions were the bond-distance functions; in this section we allow arbitrary continuously differentiable functions.

Let $I_{eq}, I_{in} \subset \Omega$ be mutually exclusive subsets of functions, and define the pair of indices together to be $I = (I_{eq}, I_{in})$. If $q_i \in \Omega$ we say it is a constraint, if $q_i \in I_{eq}$ we say it is an inequality, and we call the pair $I$
the labels. When the functions are bond-distance functions, we sometimes identify the pair \((i, j)\) with the function \(q_{ij}\), as in the setup in Section 2.1 Define
\[
M_I = \{ x \in \mathbb{R}^n : q_i(x) = 0 \text{ for } q_i \in I_{eq}, \ q_j(x) > 0 \text{ for } q_j \in I_{in} \}.
\] (2.7)

Under some mild conditions on the constraints, \(M_I\) is an open manifold. To introduce these conditions, let \(m_I = |I_{eq}|\) be the number of constraints, and construct an \(n \times m_I\) matrix whose columns are the gradients of the constraints:
\[
Q_I(x) = (\nabla q_i(x))_{i \in I_{eq}}.
\] (2.8)

If \(Q_I\) has full rank everywhere in \(M_I\), then by the implicit function theorem, \(M_I\) is an open manifold of dimension \(d_I = n - m_I\). In this case there is a natural measure \(\mu_I(dx)\) on \(M_I\), the \(d_I\)-dimensional Hausdorff measure, which is obtained from the Euclidean measure in the ambient space \(\mathbb{R}^n\) by restriction.

Define a collection of labels
\[
J = \{ I^{(1)}, I^{(2)}, \ldots, I^{(|J|)} \}.
\] (2.9)

We make the assumption that for each \(I \in J\), \(Q_I\) has full rank everywhere in \(M_I\). (Assumption 1.) In applications this assumption doesn’t always hold, and we comment further on what can happen when it doesn’t in the conclusion.

Define
\[
S = \bigcup_{I \in J} M_I.
\] (2.10)

The set \(S\) is a stratification, given some additional mild but technical conditions on the relationship between manifolds\([19],[18]\). We assume these conditions hold, and refer to \(S\) as a stratification hereafter.

We wish to sample a probability distribution on the stratification \(S\). This probability distribution is formed from a collection of scalar functions \(\{f_i(x)\}_{i \in J}\), representing the density on each manifold \(M_I\) with respect to its natural measure \(d\mu_I\), up to a global constant of proportionality. Define a probability measure by
\[
\rho(dx) = Z^{-1} \sum_{I \in J} f_i(x) \mu_i(dx), \quad Z = \sum_{I \in J} \int_{M_I} f_i(x) \mu_i(dx).
\] (2.11)

Assume that each of the integrals exists and is finite. Our Monte-Carlo method will produce samples from \(\rho(x)\).

### 2.2.2 Measure for sticky spheres

The equilibrium distribution \(\rho_I(x)\) in (2.5) for sticky spheres can be put in the form (2.11) using the co-area formula, which expresses delta-functions in terms of the natural measure on each manifold \([13],[42]\).

Define \(\kappa_I = \prod_{(i,j) \in I_{eq}} \kappa_{ij}\) to be the total stickiness of the bonds in \(I_{eq}\). Then \(\rho_I(x)\) has the form \([8]\)
\[
\rho_I(dx) = Z^{-1} \sum_{I \in J} \kappa_I |Q_I(x)|^{-1} \mu_I(dx), \quad Z = \sum_{I \in J} \int_{M_I} |Q_I(x)|^{-1} \mu_I(dx).
\] (2.12)

Here \(|A|\) for a rectangular matrix \(A\) is the pseudodeterminant of \(A\), equal to \(|A^T A|^{1/2}\) for a matrix with more rows than columns, or \(|AA^T|^{1/2}\) for a matrix with more columns than rows. Physically, the quantity \(|Q_I(x)|^{-1}\) is proportional to the vibrational entropy of \(x\) \([22],[29]\).
2.2.3 Neighbours and connectedness of the stratification

Our algorithm to sample from $\rho(x)$ in (2.11) depends on the manifolds in the stratification being connected, in a way we now describe. Roughly, we need each manifold of dimension $d < n$, to be in the closure of a manifold of dimension $d + 1$. This is usually the case when manifolds are formed by adding constraints one-by-one, since, if we add one constraint $q$ to a set $I_{eq}$, then its level set $\{x : q(x) = 0\}$ often either intersects $M_I$, or, if $q \notin I_{in}$, is on the boundary of its closure.

Specifically, given labels $I, J \in \mathcal{J}$, we say that $J$ is a Gain neighbour of $I$, and that $I$ is a Lose neighbour of $J$, if

$$I_{eq} = J_{eq} \cup \{q\}, \quad q \not\in J_{eq}. \tag{2.13}$$

When the above holds, then Assumption 1 implies that $d_J = d_I + 1$: in going from $M_I$ to $M_J$, we gain a dimension, and in going from $M_J$ to $M_I$ we lose a dimension. For each $I \in \mathcal{J}$, let the set of Gain and Lose neighbours of $I$ be, respectively,

$$N_{\text{gain}}(I), N_{\text{lose}}(I) \subset \mathcal{J}. \tag{2.14}$$

Clearly if $J \in N_{\text{gain}}(I)$, then $I \in N_{\text{lose}}(J)$.

We say that $\mathcal{J}$ is connected if there is a path between any pair $I, J \in \mathcal{J}$ by following Gain or Lose neighbours. We make the assumption that $\mathcal{J}$ is connected (Assumption 2.)

Our hope is that if $J \in N_{\text{gain}}(I)$, then $M_I \subset \partial \overline{M}_J$; that is, $M_I$ is part of the boundary of the closure of $M_J$. We will simply say that such an $M_I$ is on the boundary of $M_J$. This is a property our algorithm will rely on to pass between manifolds. Recall that for sticky spheres, if a constraint $q$ is dropped it becomes an inequality: if $I_{eq} = J_{eq} \cup \{q\}$ (disjoint union), then $q \in J_{in}$. Since the constraint $q$ changes from an inequality $q(x) > 0$ to an equality $q(x) = 0$ as we pass from $J$ to $I$, it is often the case that $M_J \subset \partial \overline{M}_J$.

If $I_{eq} = J_{eq} \cup \{q\}$ (disjoint union) and $q \in J_{in}$, we call $J$ a one-sided Gain neighbour of $I$. We also consider the more general case where $q$ is simply forgotten, so that $q \not\in J_{eq} \cup J_{in}$, we call such a $J$ a two-sided Gain neighbour of $I$. In this case it may be that $M_I \subset \overline{M}_J$; that is, $M_I$ is part of the closure of $M_J$, but not necessarily part of its boundary. In the two-sided case we will still refer to $M_I$ as being part of the boundary of $M_J$, because it may be treated as if it were a boundary in our algorithm.

2.3 Examples of stratifications

**Example 1.** (Parabola and line in two dimensions) Let $(x, y) \in \mathbb{R}^2$, and let $\Omega = \{q_1(x, y), q_2(x, y)\}$ with

$$q_1(x, y) = y - x^2, \quad q_2(x, y) = 2 - y.$$
Consider the stratification formed by considering all possible labelings of $q_1, q_2$ as constraints or inequalities: $J = I^{(1)} \cup I^{(2)} \cup I^{(3)} \cup I^{(4)}$ with $I^{(1)} = \emptyset$, $I^{(2)} = \{q_1, q_2\}$, $I^{(3)} = \{q_2, q_1\}$, $I^{(4)} = \{q_1, q_2\}$. That is, writing $M_k = M_{I(k)}$, the manifolds are defined mathematically and named for ease of reference as

- "Interior": $M_1 = \{(x, y) : y > x^2, y < 2\}$ (two-dimensional, blue)
- "Parabola": $M_2 = \{(x, y) : y = x^2, y < 2\}$ (one-dimensional, red)
- "Line": $M_3 = \{(x, y) : y = 2, y > x^2\}$ (one-dimensional, yellow)
- "Corners": $M_4 = \{(x, y) : y = x^2, y = 2\}$ (zero-dimensional, purple.)

The stratification is visualized in Figure 1, with each manifold a different color as listed above. This stratification is connected, since $M_1$ is a Gain neighbour of both $M_2, M_3$, and each of $M_2, M_3$ in turn is a Gain neighbour of $M_4$. Since each function is either a constraint or an inequality, all Gain neighbours are one-sided Gain neighbours.

Each manifold comes with a natural measure $\mu_k(dx) = \mu_{I(k)}(dx)$. The most familiar are the measures for the interior, $\mu_1(dx)$, and line, $\mu_2(dx)$, which are the two-dimensional and one-dimensional area measures on the corresponding sets respectively. The measure on the parabola, $\mu_3(dx)$, is the arc-length measure: if $A \subset M_3$ is a connected subset of the parabola, then $\int_A \mu_3(dx)$ equals the arc length of $A$. The corner’s measure $\mu_4(dx)$ is the counting measure, which is $\mu_4(A) \in \{0, 1, 2\}$ depending on how many points from the set $M_4 = \{(-\sqrt{2}, 2), (\sqrt{2}, 2)\}$ are included in $A$.

One example of a probability measure on $S$ is

$$\rho(dx) \propto \mu_1(dx) + \mu_2(dx) + \mu_3(dx) + \mu_4(dx). \quad (2.15)$$

This weights points on each manifold according to their arc-length measures. Some points sampled from this measure are shown in Figure 1; we explain how they were sampled in Section 2.

**Example 2. (A trimer of sticky discs in two dimensions)** Consider a trimer of identical two-dimensional discs with radius $r$. The three bond-distance functions are

$$q_{12}(x) = |x_1 - x_2|^2 - 4r^2, \quad q_{13}(x) = |x_1 - x_3|^2 - 4r^2, \quad q_{23}(x) = |x_2 - x_3|^2 - 4r^2,$$

and $Q = \{q_{12}, q_{23}, q_{13}\}$. Suppose we wish to sample two types of configurations: a polymer whose internal angle can change, and a triangle that can only translate and rotate. We define $J = \{I, J\}$ with $I = (I_{eq}, I_{in})$, $J = (J_{eq}, J_{in})$ and

$$I_{eq} = \{(1, 2), (2, 3)\}, \quad I_{in} = \{(1, 3)\}, \quad J_{eq} = \{(1, 2), (2, 3), (1, 3)\}, \quad J_{in} = \emptyset.$$
Figure 3: Selected configurations of a polymer of \(N = 6\) unit spheres, from Example 3. Bonds in the backbone of the polymer are fixed and cannot break, while all other bonds can break and form.

Figure 4: In Example 3 with identical sticky parameter for all bonds, the relative probabilities of forming the octahedron (left) and polytetrahedron (middle) are 5% and 95% respectively, independent of sticky parameter. For particles of types A and B as shown in the image on the right, we will show that by choosing the sticky parameters \(\kappa_{AA}, \kappa_{AB}, \kappa_{BB}\) for corresponding interacting pairs appropriately, we can make the octahedron form with nearly 100% probability.

Manifold \(M_I\) contains all the configurations of the polymer, and manifold \(M_J\) contains all the configurations of the triangle. Some of these configurations are illustrated in Figure 2. Notice that when constraint \((1, 3)\) is dropped from \(J\) to get \(I\), it becomes an inequality, so \(I\) is a one-sided Gain neighbour of \(J\).

**Example 3.** (A polymer of 6 sticky unit spheres in three dimensions) Consider a collection \(N = 6\) sticky three-dimensional spheres with unit diameter, with sticky parameter \(\kappa\) for each bond. Suppose the spheres are connected in a chain, forming a polymer, and the bonds in the backbone of the polymer cannot break. We form a stratification by letting \(Q = \{q_{ij}(x)\}_{(i,j) \in E}\), and considering all possible ways to let the non-backbone pairs come into contact:

\[ J = \{ I : \{(1, 2), (2, 3), (3, 4), (4, 5), (5, 6)\} \subset I_{eq}, I_{in} = E - I_{eq} \}. \]

Some of the labels in \(J\) are infeasible, in the sense that \(M_I = \emptyset\); for example, it is impossible to find a configuration where all pairs of spheres are in contact. The minimum number of bonds of a feasible configuration in \(J\) is 5, and the maximum number is 12. Some example configurations are shown in Figure 3.

To form a probability measure we must choose the strengths of the interactions that are not in the backbone. We consider two possibilities. One is where all bonds have equal sticky parameter \(\kappa\), so the equilibrium distribution is

\[ \rho_{\text{equal}}(dx) = Z^{-1} \sum_{I \in J} k^{\|I\| - |J|} |Q_I(x)|^{-1} \mu_I(dx). \]  

(2.16)
Figure 5: Configurations drawn from the equilibrium distribution of a freely-jointed polymer (left) and a semiflexible polymer (right) with $N = 20$ spheres, with sticky parameter $\kappa = 1$ with the wall and with bending stiffness parameter $k_{\text{bend}} = 2$ in the semiflexible case. Blue spheres are in contact with the wall and green spheres are in free space.

Because the equilibrium distribution only depends on the total number of bonds in a cluster, the relative probabilities between clusters with the same number of bonds is always the same, independent of sticky parameter.

We might wish to control which cluster occurs with highest probability, a problem that arises when studying the self-assembly of colloids \cite{26, 58}. Therefore, we also consider a measure formed from imagining that spheres 2,3,4,5 are of type “A”, and that spheres 1,6 are of type “B”, with sticky parameters for AA, AB, BB interactions equal to $\kappa_{AA}, \kappa_{AB}, \kappa_{BB}$ respectively, as illustrated in Figure 4. The equilibrium distribution is

$$\rho_{AB}(dx) = Z^{-1} \sum_{I \in I} \kappa_{AA}^{n_{AA}(I)} \kappa_{AB}^{n_{AB}(I)} \kappa_{BB}^{n_{BB}(I)} |Q_I(x)|^{-1} \mu(dx).$$

Example 4. (Polymer adsorbing to a surface) A classic problem in polymer physics is to understand the statistical mechanics of a polymer that can adsorb weakly to a surface \cite{10, 12}. As the strength of interaction with the surface increases, the polymer behaves less like a three-dimensional polymer and more like a two-dimensional polymer (e.g. \cite{40}, and references therein.) We can study this problem in the limit when the range of interaction with the surface is very short, so the particles are sticky on the surface. Consider a polymer formed from $N$ identical spheres with unit diameter that adsorb to a flat surface. Define a collection of functions $\Omega = \Omega_{\text{bonds}} \cup \Omega_{\text{surf}}$, with $\Omega_{\text{bonds}} = \{q_{i,i+1}(x)\}_{i=1...N-1}$ where $q_{i,i+1}(x) = |x_i - x_{i+1}|^2 - 1$, and $\Omega_{\text{surf}} = \{w_i(x)\}_{i=1...N}$ where $w_i(x) = x_{i3}$. We form a stratification by considering all possible ways for the spheres to stick to the surface, assuming the first sphere is always constrained to it:

$$J = \{I : I_{\text{eq}} = \Omega_{\text{bonds}} \cup \Omega_{\text{surf}} \text{ where } w_1 \in \Omega_{\text{surf}} \subset \Omega_{\text{surf}}, \ I_{\text{in}} = \Omega_{\text{surf}} - \Omega_{\text{surf}} \}.$$  

For simplicity we don’t consider inequalities between spheres in this example, though one could easily include them.

We will study a freely-jointed polymer, whose only energy comes from interactions with the surface. Given sticky parameter $\kappa$ with the wall, the equilibrium distribution is

$$\rho_{\text{free}}(dx) = Z^{-1} \sum_{I \in J} \kappa_{\text{surf}}^{|I_{\text{surf}}|} |Q_I(x)|^{-1} \mu(dx).$$

We will also study a semiflexible polymer, which has a bending energy that penalizes deviations from a straight line:

$$\rho_{\text{bend}}(dx) = Z^{-1} e^{-\sum_{i=1}^{N-2} \frac{1}{2} k_{\text{bend}} (1 - \cos \theta)} \sum_{I \in J} \kappa_{\text{surf}}^{|I_{\text{surf}}|} |Q_I(x)|^{-1} \mu(dx).$$
Figure 6: Example 5, with \( n = 3 \). To estimate the surface area of the ellipse (grey), we form a stratification by intersecting it first with the blue plane, then with the red plane. The entire set of surfaces intersect in only two points, whose union is a 0-dimensional manifold. Since the Hausdorff volume of this 0-dimensional manifold is a known value (it is 2), we can infer the volumes of the other manifolds in the stratification from points sampled within the entire stratification. Later, we will apply this strategy to calculate the surface area of a 10-dimensional ellipsoid.

Here \( k_{\text{bend}} \) is a parameter measuring the bending stiffness, and \( \cos \theta_i = (x_{i+2} - x_{i+1}) \cdot (x_{i+1} - x_i) \) is cosine of the \( i \)th internal angle \( \theta_i \) in the polymer.

**Example 5. (Surface area of an \( n \)-dimensional ellipsoid)** Our final example illustrates how to use the Stratification Sampler to estimate the volume or surface area of a manifold. When the intrinsic dimension of the manifold is high enough (roughly larger than 4), deterministic methods cannot typically be used to compute its volume, because they require too many points to adequately cover the manifold \[50]. Consider the surface of an \( n \)-dimensional ellipsoid, defined as the solution to

\[
E_n = \{ x \in \mathbb{R}^n : q_1(x) = \frac{x_1^2}{a_1^2} + \frac{x_2^2}{a_2^2} + \ldots + \frac{x_n^2}{a_n^2} - 1 = 0 \}.
\] (2.20)

Here \( a_1, \ldots, a_n > 0 \) are the lengths of the semiaxes. Let \( \text{Vol}(E_n) \) be the \( n-1 \)-dimensional volume of \( E_n \) with respect to the natural Hausdorff measure; we call this simply the surface area of \( E_n \). While there exist expressions for the surface area of \( E_n \), they are complicated (see Equation (10) in \[45]\)), so we show instead how one can estimate the surface area through sampling.

The idea is to construct a stratification which contains at least one manifold whose volume we know, or can easily estimate. Since we can exactly calculate the volume of a 0-dimensional manifold, by counting the number of points in it, our strategy will be to add constraints until the intersection of all the constraints is a collection of isolated points. To this end, let

\[
\Omega = \{ q_1(x), q_2(x) = x_2, q_3(x) = x_3, \ldots, q_n(x) = x_n \}.
\]

Let \( I^{(k)} = (I^{(k)}_{\text{eq}}, I^{(k)}_{\text{in}}) \) with \( I^{(k)}_{\text{eq}} = \{ q_1, q_2, \ldots, q_k \}, I^{(k)}_{\text{in}} = \emptyset \). We form a stratification from

\[
J = \bigcup_{k=1}^{n} I^{(k)}.
\]

That is, we intersect the ellipsoid with an ordered collection of planes going through the origin. See Figure 6 for an illustration of the stratification when \( n = 3 \).

We define a measure on the stratification induced by \( J \) from the Hausdorff measures \( \mu_k = \mu_{I^{(k)}} \) on each manifold \( M_k = M_{I^{(k)}} \) as

\[
\rho_E(dx) = Z^{-1} \sum_{k=1}^{n} c_k \mu_k(dx).
\] (2.21)
Here \( c_k \in \mathbb{R}^+ \) is a constant weight for each manifold, and \( Z \) is the normalizing constant. A good strategy is to choose the weights \( \{c_k\} \) so the sampler spends roughly the same amount of time in each manifold. Such a strategy has been shown to be optimal in certain systems when temperature is the variable that changes, rather than dimension [38].

If we have a collection of points \( X_1, X_2, \ldots, X_m \sim \rho_x \), then we can estimate the surface area of \( E_n \) as

\[
\text{Vol}(E_n) \approx \frac{\text{# of points in } M_1}{\text{# of points in } M_n} \cdot \frac{2c_n}{c_1}
\]  

(2.22)

This estimate comes from observing that, if the algorithm is ergodic, then

\[
\frac{\sum_{i=1}^m 1_{M_1}(X_i)}{\sum_{i=1}^m 1_{M_n}(X_i)} \to \frac{\int_{M_1} \rho(d x)}{\int_{M_n} \rho(d x)} = \frac{\int_{M_1} c_1 \mu_1(d x)}{\int_{M_n} c_n \mu_n(d x)}
\]

as \( m \to \infty \). Since \( \int_{M_1} c_1 \mu_1(d x) = c_1 \text{Vol}(E_n) \), and \( \int_{M_n} c_n \mu_n(d x) = 2c_n \) since the manifold \( M_n = \{ \pm a_1 \} \) contains exactly two points, we solve for \( \text{Vol}(E_n) \) to get the estimate above.

3 Algorithm

3.1 Overview

We present an algorithm for generating a Markov chain \( X_1, X_2, \ldots \in S \) with invariant distribution \( \rho \), defined in (2.11). We call this algorithm the Stratification sampler. The algorithm keeps track of both the point and its label as a pair \( (X_k, l_k) \). Given \( (X_k, l_k) \) with \( X_k = x \in M_{l_k}, l_k = I \), the next step \( (X_{k+1}, l_{k+1}) \) of the Markov chain is generated by first constructing a proposal move \((y, J)\), and then accepting or rejecting it using a Metropolis-Hastings step.

The proposal move is constructed in two steps:

(i) Choose a label \( J \) with probability \( \lambda_{IJ}(x) \);
(ii) Choose a point \( y \in M_J \) with probability density \( h_{IJ}(y|x) \) with respect to \( \mu_J(d x) \).

Here \( \lambda_{IJ}(x) \) is the probability of proposing a label \( J \), given the current label is \( I \) and the current point is \( x \). Since it is a probability distribution on labels, we must choose this function for each \( (x, I) \) so that \( \sum_{J \in \mathcal{J}} \lambda_{IJ}(x) \leq 1 \). The function \( h_{IJ}(y|x) \) is the probability density of proposing a point \( y \in M_J \), given the current label is \( I \), the current point is \( x \), and the proposed label is \( J \). Sometimes the method fails to produce a proposal point \( y \), in which case the proposal is set to \( (x, I) \). Therefore it is usually the case that \( \int_{M_I} h_{IJ}(y|x) \mu_J(d y) < 1 \).

Given a proposal \((y, J)\), it is accepted according to a Metropolis-Hastings rule with acceptance probability

\[
a(y, J|x, I) = \min \left( 1, \frac{f_J(y) \lambda_{JI}(y) h_{JI}(x|y)}{f_I(x) \lambda_{IJ}(x) h_{IJ}(y|x)} \right).
\]

(3.1)

That is, the algorithm generates \( U \sim \text{Unif}([0, 1]) \), and if \( U < a(y, J|x, I) \) the proposal is accepted and the next step is \( X_{k+1} = y, l_{k+1} = J \). Otherwise, the proposal is rejected and the next step is \( X_{k+1} = x, l_{k+1} = I \). If the proposal itself was \((x, I)\), it is accepted automatically. Importantly, as with most Monte-Carlo samplers, this method can be implemented without knowing the overall normalization constant \( Z \), since \( Z \) cancels out in the acceptance ratio above.

We show in Appendix A that if \( X_k \sim \rho \), then \( X_{k+1} \sim \rho \). Therefore \( \rho \) is an invariant measure for the Markov chain above. We expect that for reasonable choices of proposals the algorithm is ergodic, so that for any initial condition, \( X_0 \to \rho \) as \( n \to \infty \).

The choice of label-change probabilities \( \lambda_{IJ}(x) \) and densities \( h_{IJ}(y|x) \) are critical to implementing the method efficiently. Given a point \((x, I)\) and its proposal \((y, J)\), we must be able to evaluate the density
These probabilities are set so that Gain or Lose move are obtained from the individual label proposal probabilities as

\[
\text{In the second two cases the inequalities may vary arbitrarily. The total probabilities of choosing a Same, the specific label proposal densities in Section 3.3.}
\]

The total probabilities of choosing a Same, Move types, and we give the formula for proposal densities. We also explain how to calculate the proposal normal space. Under Assumption 1, \(\dim(T_x)\) is neither too high nor too low. The average acceptance probability should be sufficiently large that computation isn’t wasted generating proposal moves. However, if it is too large, successive samples are highly correlated, so many samples must be generated to obtain approximately independent draws from \(\rho\).

We explain our choices of \(\lambda_{ij}(x), h_{ij}(y|x)\) in Sections 3.2 and 3.3. Section 3.2 is focused on the geometry of the move types, while Section 3.3 describes specific proposal densities. The overall algorithm is summarized in pseudocode in Appendix B, and codes are available at [1].

### 3.2 Proposal moves

We consider three different types of label proposals, that we call Same, Gain, Lose:

- **Same**: choose \(J = I\).
  
  This retains the current labels, and takes a step on the current manifold.
- **Gain**: choose \(J \in N_{\text{gain}}(I)\).
  
  This moves to a manifold of one higher dimension, obtained by dropping a single constraint.
- **Lose**: choose \(J \in N_{\text{lose}}(I)\).
  
  This moves to a manifold of one lower dimension, obtained by adding a single constraint.

In the second two cases the inequalities may vary arbitrarily. The total probabilities of choosing a Same, Gain or Lose move are obtained from the individual label proposal probabilities as

\[
\begin{align*}
\Lambda_{\text{same}}(x) &= \lambda_{11}(x), \\
\Lambda_{\text{gain}}(x) &= \sum_{J \in N_{\text{gain}}(I)} \lambda_{1J}(x), \\
\Lambda_{\text{lose}}(x) &= \sum_{J \in N_{\text{lose}}(I)} \lambda_{1J}(x). 
\end{align*}
\]

These probabilities are set so that \(\Lambda_{\text{same}}(x) + \Lambda_{\text{gain}}(x) + \Lambda_{\text{lose}}(x) = 1\) for each \(x\). We describe how to choose the specific label proposal densities in Section 3.3.

Given a proposal \(J\), we then choose \(y \in M_J\). We now describe how to do this for each of the three move types, and we give the formula for proposal densities. We also explain how to calculate the proposal densities for the reverse move, \(y \to x\), when all we are given is \(x, y\); this calculation is required to evaluate the acceptance probability (3.1).

In what follows, we define \(T_{x,I}\) to be the tangent space to \(M_I\) at \(x \in M_I\), and \(N_{x,I} = T_{x,I}^\perp\) to be the normal space. Under Assumption 1, \(\dim(T_{x,I}) = d_I\), \(\dim(N_{x,I}) = m_I\). We let \(T_{x,I} \in \mathbb{R}^{n \times d_I}\) be a matrix whose columns form an orthonormal basis of \(T_{x,I}\). For a given vector \(v \in T_{x,I}\), we let \(T_{x,I,v} = T_{x,I} \cap (\text{span}\{v\})^\perp\); this is the \(d_I-1\)-dimensional subspace of the tangent space \(T_{x,I}\) that is orthogonal to \(v\). Let \(T_{x,I,v} \in \mathbb{R}^{n \times d-I}\) be a matrix whose columns form an orthonormal basis of \(T_{x,I,v}\). Matrices of the form \(T_{x,I}, T_{x,I,v}\) can be calculated from matrices of the form \(Q_I\) in (2.8) using standard linear algebra operations; we use the QR decomposition as described in Appendix B, Algorithm 5.

#### 3.2.1 Same move: \(x \in M_I, y \in M_I\)

**Generating the move** To generate a proposal on \(M_I\), we use the method proposed in [57], which works as follows. We first choose \(v \in T_{x,I}\) according to a density \(p_{\text{same}}(v|x)\). We give examples of such densities in Section 3.3. Then, we find \(y \in M_I\) such that \(y - (x + v) \in N_{x,I}\). Since by Assumption 1 the columns of \(Q_I(x)\) span \(N_{x,I}\), we may find \(y\) by solving the system of equations

\[
q_i(x + v + Q_I(x)a) = 0, \quad i \in I_{\text{eq}},
\]

for the unknown \(a \in \mathbb{R}^{m_I}\). This may be done numerically using a nonlinear equation solver, such as Newton’s method. Our specific implementation is given in Appendix B, Algorithm 4. Whatever solver
is used must be deterministic with a deterministic initial condition as we explain momentarily. Let NES denote the particular choice of nonlinear equation solver used in the algorithm.

If a solution $a$ is successfully found by the NES, the proposal is constructed from the solution $a$ as $y = x + v + Q_i(x)a$. It must then be checked that $y$ satisfies the inequalities in $I_{eq}$; if not, the proposal is $y = x$. Additionally, if the solver fails to find a solution, the proposal is $y = x$. The solver could fail for a variety of reasons: there could be no solution for a particular $v$, or, even if there is a solution, the NES could simply fail to converge to a solution.

The density for a successful proposal that is found by the NES and satisfies the inequalities is $p_{II}^{\text{same}}(v;x)\frac{\partial y}{\partial x}$, where $\frac{\partial y}{\partial x}$ is the inverse of the determinant of the Jacobian of the transformation from $v \to y$. This was shown in [57] to be $|T_{s,I}^T T_{y,I}|$. For a plane, $|\frac{\partial y}{\partial x}| = 1$, since $T_{s,I} = T_{y,I}$.

Let $A_{\text{same}}(x)$ be set of values of $y$ that are accessible from $x$ via a Same move: they can be found by solving (3.3) using our NES; by definition they satisfy the inequalities. In order for $A_{\text{same}}(x)$ to be well-defined, we must use a deterministic initial condition for the NES. In addition, there must be only one $v$ for each move $x \to y$, a fact we verify shortly. Let $1_{A_{\text{same}}}(x)(y)$ be the characteristic function for the sets $A_{\text{same}}(x)$, equal to 1 if $y \in A_{\text{same}}(x)$, and 0 otherwise. The proposal density for a successful proposal is

$$h_{II}(y|x) = p_{II}^{\text{same}}(v;x)|T_{s,I}^T T_{y,I}|1_{A_{\text{same}}(x)}(y).$$ (3.4)

**Evaluating the density for the reverse move**

Given $x, y \in M_I$, we now explain how to evaluate the proposal density (3.4). We focus on evaluating the density from $x \to y$, to avoid rewriting (3.4), but in practice one proposes a $y$ and then evaluates the density for the move from $y \to x$, and uses this density to calculate the acceptance probability in (5.4).

The step $v$ can be found by projecting $y - x$ onto $T_{s,I}$, as

$$v = T_{s,I} T_{y,I}^T (y - x).$$ (3.5)

From this we can evaluate $p(v;x)$. Notice that, as required, $v$ is uniquely defined from $x, y$.

We also need to evaluate $1_{A_{\text{same}}(x)}(y)$. To do this, we must run the NES starting from $x$ with tangent step $v$, and check two things: (i) the NES produces a solution, and (ii) this solution equals $y$. Even though we know that $y$ is a solution to (3.3), because the pair $(x,y)$ was a successful proposal in a forward move, we still need to check that the NES actually produces this solution. If not, the density for $y$ must be set to zero.

Therefore, the NES must be run twice for each move: once to produce a forward move, and once to compute the density of the reverse move. That this check is necessary for detailed balance was first pointed out in [57], and [35] showed empirically that samplers can be more efficient for proposals where this check is critical to computing the correct acceptance probability.

**3.2.2 Gain move: $x \in M_I, y \in M_J$ with $J \in N_{\text{gain}}(I)$**

**Generating the move**

A Gain move is very similar to a Same move. Suppose that $q$ is the constraint dropped from $I_{eq}$ to obtain $J_{eq}$. We have $x \in M_I$, and we wish to propose $y \in M_J$ with $J_{eq} = I_{eq} \setminus \{q\}$.

Since $x \in M_I$, the closure of $M_J$ (we know this since $q(x) = 0$), this can be done in exactly the same way as for a Same move, except the move is on $M_J$, not $M_I$. If a proposal fails, either due to inequality constraints, or because the NES fails, then we set $y = x, J = I$.

**Density of the proposal**

Let $A_{\text{gain}}(x)$ be the set of values of $y$ that are accessible from $x$ via a Gain move using our NES; by definition they satisfy the inequalities in $J_{in}$. The proposal density is

$$h_{II}(y|x) = p_{II}^{\text{gain}}(v;x)|T_{s,I}^T T_{y,J}|1_{A_{\text{gain}}(x)}(y).$$ (3.6)
Evaluating the density for the reverse move  Given $x \in M_I$, $y \in M_J$, the density (3.6) is evaluated in the same way as for a Same move: one first computes $v \in T_{x,J}$, and then one runs the NES to check whether $y \in A_{\text{gain}}(x)$.

3.2.3 Lose move

Generating the move  Suppose that $q$ is the constraint added to $I_{\text{eq}}$ to obtain $J_{\text{eq}}$. We have $x \in M_I$, and we wish to propose $y \in M_J$ with $J_{\text{eq}} = I_{\text{eq}} \cup \{q\}$.

We first choose a unit vector in the tangent plane, $v \in T_{x,J} \cap B_*(1)$, where $B_*(1)$ is the surface of the unit sphere centered at $x$. The density for this choice is $p_{IJ}^{\text{lose}}(v; x)$. Constructing a density on the unit sphere whose normalization constant is known analytically can be hard, but we explain some choices in Section 3.3. This unit vector gives the direction to step in the tangent plane, but the magnitude of the tangent step is an unknown variable $\alpha \in \mathbb{R}$.

Next, we find $y \in M_J$ such that $y - (x + \alpha v) \in N_{x,J}$. This is done by solving the system of equations

$$q_j(x + \alpha v + Q_j(x)\alpha) = 0, \quad j \in J_{\text{eq}},$$

for the unknown $(a, \alpha) \in \mathbb{R}^{m_I}$. There are $m_J$ equations and $m_I$ unknowns, so this system is well-posed in general. Our specific method to solve using Newton’s method is given in Appendix B.

In general, call the method used to solve (3.7) NES-L. As before this method must be deterministic with a deterministic initial condition.

If the solver NES-L finds a solution $(a, \alpha)$, and if $\alpha > 0$, then the proposal is $y = x + \alpha v + Q_j(x)\alpha$. If the reconstructed $y$ fails to satisfy the inequalities in $J_{\text{eq}}$, or if NES-L fails to find a solution, the proposal is rejected and set to $y = x, J = I$.

We reject the proposal when $\alpha < 0$ in order to ensure reproducibility for the reverse move. If for some $v$ (3.7) has solution $(a, \alpha)$, then the solution for $-v$ is $(a, -\alpha)$. If we are given only $x, y$, then we don’t know which of $v, -v$ was used in the proposal, so without a sign restriction on $\alpha$ we can’t evaluate the density of that move, nor whether $y$ is accessible from $x$.

This is also the reason why we choose a unit vector for $v$, not an arbitrary vector. If we chose an arbitrary vector, we could still solve (3.7), but then given only $x, y$ we would not know which $v$ was actually used in the proposal.

Density of the proposal  Let $A_{\text{lose}}(x) \subset M_J$ be the set of values of $y$ that are accessible from $x$ via a Lose move using our NES; these necessarily satisfy the inequalities in $J_{\text{eq}}$. The density of a successful proposal $y \in A_{\text{lose}}(x)$ is $p_{IJ}^{\text{lose}}(v; x)\left|\frac{\partial v}{\partial y}\right|$, where as before $\left|\frac{\partial v}{\partial y}\right|$ is the determinant of the inverse of the Jacobian of the transformation from $v \rightarrow y$; the difference from the previous two move types is that now $v$ is constrained to be a unit vector. Therefore, to linear order $v$ can only vary in the subspace $T_{x,J,v}$.

The Jacobian factor can be calculated to be

$$\left|\frac{\partial v}{\partial y}\right| = \alpha^{-d_j}|T_{x,J,v}T_{y,J}|,$$

where $\alpha = |y - x|$. Therefore the proposal density is

$$h_{ij}(y|x) = p^{\text{lose}}_{IJ}(v; x)|T_{x,J,v}T_{y,J}| |y - x|^{-d_j}1_{A_{\text{lose}}}(x)(y).$$

To calculate the Jacobian, suppose that $y, v$ vary as $y \rightarrow y + \Delta y, v \rightarrow v + \Delta v$, and simultaneously $\alpha \rightarrow \alpha + \Delta \alpha$. To linear order we must have $\Delta y = T_{x,J,b}$ for some $b \in \mathbb{R}^{d_j}$, $\Delta v = T_{x,J,c}$ for $c \in \mathbb{R}^{d_I - 1} = \mathbb{R}^{d_I}$. The Jacobian $\frac{\partial v}{\partial y}$ will be the matrix $A$ such that to linear order $c = Ab$. Linearizing the equation $y = x + \alpha v + w$ with $w \perp T_{x,J}$ gives

$$\Delta y = \alpha \Delta v + v \Delta \alpha + \Delta w,$$

where $\Delta w \perp T_{x,J}$. Multiplying the above equation by $T_{x,J,v}$, using that $T_{x,J,w} = 0$, $T_{x,J,v}v = 0$, and substituting the expressions for $\Delta y, \Delta v$ gives

$$T_{x,J,v}T_{y,J}b = ac \quad \Leftrightarrow \quad c = a^{-1}T_{x,J,v}T_{y,J}b.$$

The Jacobian factor is therefore $|\alpha^{-1}T_{x,J,v}T_{y,J}|$. 

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is added to $\Lambda$ with standard deviation $R$ constructed from a vector of independent random variables $N$.

or Lose, the particular label $J$ the type of move is chosen with probabilities $\lambda$.

Evaluating the density for the reverse move

Given $x \in M_I$, $y \in M_I$ we find $v$ by first calculating $\bar{v} = T_{x,I} T_{y,I}^T(y - x)$, and then setting $v = \bar{v} / |\bar{v}|$, $a = |\bar{v}|$. We must also check that $y \in A_{\text{lose}}(x)$, by running the NES-L to solve (3.7) with the calculated $v$. In addition to checking that the solver converges, and gives $y$ as a solution, we must also check that the solution $(\alpha', a')$ has $\alpha' > 0$.

**3.3 Specific proposal densities**

A key part of implementing this method is choosing the proposal densities $\lambda_{ij}(x)$, $p_{ij}^*(v; x)$. Section 3.3.1 describes the simplest possible choice, where all proposals are isotropic; this turns out to be too inefficient to use in practice. Section 3.3.2 describes a more complex choice, that treats directions pointing towards and away from the boundaries anisotropically; this choice is very efficient in practice.

**3.3.1 Simplest choice of proposals: isotropic Gain and Lose moves**

To choose the type of move, we define parameters $\lambda_{\text{gain}}, \lambda_{\text{lose}}$ and calculate $\lambda_{\text{same}} = 1 - \lambda_{\text{gain}} - \lambda_{\text{lose}}$, and set the overall probabilities of each move type in (3.2) as

$$
\lambda_{\text{gain}}(x) = \lambda_{\text{gain}}, \quad \lambda_{\text{lose}}(x) = \lambda_{\text{lose}}, \quad \lambda_{\text{same}}(x) = \lambda_{\text{same}}.
$$

The type of move is chosen with probabilities $\lambda_{\text{gain}}(x), \lambda_{\text{lose}}(x), \lambda_{\text{same}}(x)$, and if the proposed move is Gain or Lose, the particular label $J$ is chosen uniformly among the neighbours $N_{\text{gain}}(I)$ or $N_{\text{lose}}(I)$. If either of $N_{\text{gain}}(I), N_{\text{lose}}(I)$, is empty, the corresponding probability is set to $\lambda_{\text{same}}(x) = 0$, and the leftover probability is added to $\lambda_{\text{same}}(x)$.

For a Same move from $x \in M_I$, we choose the tangent step $V \in T_{x,I}$ to be an isotropic Gaussian with standard deviation $\sigma$, which for simplicity we present as being independent of the manifold. This is constructed from a vector of independent random variables $R \in \mathbb{R}^{d_I}$ as

$$
V = T_{x,I} R, \quad R_i \sim N(0, \sigma^2), \quad i = 1, \ldots, d_I.
$$

Figure 7: Acceptance probabilities for the simple isotropic proposals in Section 3.3.1 for a Lose move from $x = (0, -k\sigma)$ to the line $\{(x_1, x_2) : x_2 = 0\}$. Here $\sigma$ is the standard deviation of the step size in the Gain proposal (3.11), and $k > 0$ controls the distance to the boundary. We chose 8 directions for a Lose proposal with vertical angles equally spaced in $\theta \in [0.05, 1.45]$ (dashed arrows), and for each proposed move (blue stars) we evaluated the acceptance probability in (3.1) with $\lambda_{\text{lose}} = \lambda_{\text{gain}}$ and $k = 1, 4$, using the formula $a(y|x) = \min(1, e^{-\frac{k}{2\sigma^2}} \sqrt{2\pi k / \cos^2 \theta})$ to be derived in (5.7). As $k$ increases, most directions give small acceptance probabilities, because they result in large displacements which are unlikely to have been proposed in a Gain move. Furthermore, such large moves are more likely to cause the NES-L to fail, before even reaching the Metropolis step. Hence, Lose moves for this kind of proposal are accepted only if they jump to a nearby boundary, i.e. if $k$ is small and the proposed direction is nearly perpendicular to the boundary. However, for nearby boundaries the corresponding Gain move is small, so has only a small probability of being accepted.
The density for such a move is

$$p_{\text{same}}(v; x) = \frac{1}{\sqrt{(2\pi\sigma^2)^d}} e^{-\frac{|v|^2}{2\sigma^2}}.$$  \hfill (3.11)

For Gain and Lose moves, we initially tried the simplest possible proposals: for a Gain move, we chose $v$ as for a Same move, so $p_{\text{gain}}(v; x)$ also had the form (3.11). For a Lose move, we chose a direction uniformly on the unit sphere, so its density was

$$p_{\text{lose}}(v; x) = \frac{1}{\text{Area of } d_{d-1}\text{-dimensional ball}}.$$  \hfill (3.12)

While this method works, in the sense that it samples the correct probability measure, it is not very efficient. The rejection rates for Gain and Lose moves are both high, and cannot be made small by an appropriate choice of parameters. For example, for Example 3 with $\kappa = 2$, and sampling parameters $\lambda_{\text{gain}} = \lambda_{\text{lose}} = 0.4$, $\sigma = 0.3$, about 88% of Lose moves and 93% of Gain moves were rejected. These numbers were relatively insensitive to $\lambda_{\text{gain}}$, $\lambda_{\text{lose}}$ and $\sigma$, increasing somewhat for $\sigma \lesssim 0.05$. Among the Lose proposals, 63% were rejected during the NES-L step (26% because of NES-L failure, 37% because of the sign of $\alpha$), and 20% during the Metropolis step; the remaining small number were rejected during the inequality check (5%) and the reverse check (0.05%). Among the Gain proposals, 56% were rejected during the inequality check, 27% during the Metropolis step, and 9% during the NES step; the remaining 0.5% were rejected during the reverse check. Compare these statistics to those for Same moves, which are constructed in the same way as Gain moves, for which 19%, 11%, 6% were rejected for the inequality, Metropolis, and NES steps respectively.

The rejection rates for Gain and Lose moves are high because these proposals are highly asymmetric with each other: by choosing a direction isotropically, Lose moves usually propose to jump to boundaries that are far away. Not only does this make the NES-L more likely to fail (both because the boundary is far away, and because the direction proposed is not always consistent with the required sign of $\alpha$), but, even if the NES-L is successful, such moves are unlikely to have been proposed in the Gain step, hence, lead to small Metropolis factors, as illustrated in Figure 7. This is why most Lose moves are rejected in either the proposal step or the Metropolis step. Furthermore, because Gain moves don’t distinguish between directions which move to the interior of the manifold, and directions which move away from it, where the inequality is violated, about half of the Gain moves are expected to be rejected because of the inequality constraint, consistent with our empirical observations.

Proposing moves that are so unlikely to be accepted wastes computation; a sequence of smaller moves that are each rather likely to be accepted would give a more efficient sampler than one that proposes medium-sized moves that are unlikely to be accepted. In the next section we construct proposals that give a high acceptance probability over a range of conditions.

### 3.3.2 Improved proposals: anisotropic Gain and Lose moves

We introduce improved proposal densities for Gain/Lose moves and label changes, which exploit anisotropy to be more symmetric with each other, and hence to have a higher chance of being accepted. In Section 5 we show this choice of densities gives an acceptance probability of $a(y, J|x, I) = 1$ for two manifolds defined by affine constraints with $f(x) = \text{cst}$ and no inequalities. Therefore, when the step sizes for the proposals are small, we expect to have high acceptance probabilities for curved manifolds as well.

Here is an overview of the strategy for producing proposals:

- **Lose moves** only jump to boundaries that are within a certain (estimated) distance $\sigma_{\text{bdy}}$ from the current point $x$. The distance is estimated by linearizing the constraints.

- **Lose moves** construct tangent steps $V$ by choosing a deterministic component $v_{\text{opt}}$ in the direction normal to the boundary it is attempting to move to, and a random amount $R$ in the directions tangent to the boundary.
normal direction to the boundary, and to obtain \( v \in \mathbb{R}^n \) a unit vector \( V \) are sufficiently close. Consider a pair of manifolds \( I \) is done by estimating the distance to each possible boundary, and only proposing moves to boundaries that are no more than an (estimated) distance of \( \sigma \). Lose moves only attempt to jump to boundaries that are no more than an (estimated) distance of \( \sigma \). Figure 8 shows a schematic of the tangent step proposals.

Figure 8: Schematic illustrating the Gain and Lose tangent step proposals in Section 3.3.2, for a one-sided boundary \( q(x) = 0 \) in \( \mathbb{R}^2 \). Gain moves propose a step \( V \) with components \( V_n \sim \text{Unif}(0, \sigma_{\text{bdy}}) \) in the normal direction to the boundary, and \( V_t \sim \mathcal{N}(0, \sigma_{\text{tan}}^2 V_n^2) \) in the tangential direction. Lose moves propose a unit vector \( V \) by summing the vector \( v_{\text{opt}} \) in the (estimated) normal direction to the boundary, and a random amount \( t \), which increases with the magnitude of the proposed move. The improved proposals start by constructing a more restrictive set of neighbours for a Lose move. This is done by estimating the distance to each possible boundary, and only proposing moves to boundaries that are sufficiently close. Consider a pair of manifolds \( I, J \) with \( J \in \mathcal{N}_{\text{lose}}(I) \), and \( J_{\text{eq}} = I_{\text{eq}} \cup \{b\} \). The distance \( h \) to the boundary \( \{x \in M: q(x) = 0\} \), in the direction of \( v \) is estimated by linearizing the equation \( q(x + hv) = 0 \) about \( x \), to obtain

\[
q(x) + hv \cdot \nabla q = 0 \quad \Rightarrow \quad h = -\frac{q(x)}{v \cdot \nabla q(x)}. \tag{3.13}
\]

The distance \( h \) above can be positive or negative. The minimum positive value of \( h \) over all \( v \in \mathcal{T}_{x,I} \cap B_x(1) \) occurs at

\[
v_{\text{opt}} = -\text{sgn}(q(x)) \frac{P_{x,I} \nabla q(x)}{|P_{x,I} \nabla q(x)|}, \quad \text{where} \quad P_{x,I} = T_{x,I} T_{x,I}^T
\]

is the orthogonal projection matrix onto \( \mathcal{T}_{x,I} \). The minimum positive distance is then

\[
h_{\text{opt}} = \frac{|q(x)||T_{x,I} T_{x,I}^T \nabla q|}{|T_{x,I}^T \nabla q|^2} = \frac{|q(x)|}{|T_{x,I}^T \nabla q|}. \tag{3.15}
\]

We simplified this expression using that \( |T_{x,I} T_{x,I}^T \nabla q| = |T_{x,I}^T \nabla q| \), since \( T_{x,I} \) has orthonormal columns. When we need to clarify what the optimal direction and distance depend on we write \( h = h_{\text{opt}}(x, I, q) \), \( v_{\text{opt}} = v_{\text{opt}}(x, I, q) \).

Let \( \sigma_{\text{bdy}} \) be a parameter, and construct a more restrictive set of Lose neighbours at point \( (x, I) \) as

\[
\mathcal{N}_{\text{lose}}(I, x) = \{J \in \mathcal{N}_{\text{lose}}(I) : h_{\text{opt}}(x, I, q_{I\to J}) < \sigma_{\text{bdy}}\}, \tag{3.16}
\]

where \( q_{I\to J} \) is the constraint that is added in going from \( I \) to \( J \). We may now describe our improved proposals, which depend on the parameter \( \sigma_{\text{bdy}} \) as well as other parameters that we introduce in turn.

---

\(^3\)Actually one should solve \( q(x + hv + w) \) with \( w \in \mathcal{T}_{x,I} \) and \( q_i(x + hv + w) = 0 \) for \( i \in I_{\text{eq}} \), but one can show that to linear order, \( w = 0 \). Briefly: linearizing the constraints in \( I_{\text{eq}} \) gives \( \sum_{i \in I_{\text{eq}}} w \cdot \nabla q_i = 0 \), since \( q_i(x+0) = 0 \). This is in contradiction with \( w \in \mathcal{T}_{x,I} \).
Label changes We propose a Lose move only to boundaries that are estimated to be within a distance of $\sigma_{\text{bdy}}$ of the current point $(x, I)$. To this end, define
\[ n_{\text{gain}}(I) = |N_{\text{gain}}(I)|, \quad n_{\text{lose}}(I, x) = |N_{\text{lose}}^\text{bdy}(I, x)| \]
(3.17)
to be the number of Gain and Lose neighbours of $(x, I)$; we will write these simply as $n_{\text{gain}}, n_{\text{lose}}$ when it is clear which point they depend on.

Choose a parameter $\lambda_{\text{lose}}$ to be the probability of proposing a Lose move when we are sufficiently close to a boundary, and choose the probability of a Gain move to be
\[ \lambda_{\text{gain}} = \sigma_{\text{bdy}} \lambda_{\text{lose}}. \]
(3.18)
The parameters must satisfy $\lambda_{\text{lose}} < 1/(1 + \sigma_{\text{bdy}})$ to be interpreted as probabilities. If it is known that for manifolds $M_I, M_J$, the function we are sampling is $f(x) \approx f_I$, a constant, for $x \in M_I$ and $f(x) \approx f_J$, a constant, for $x \in M_J$, then we should instead choose $\lambda_{\text{gain}} = (f_J/f_I)\sigma_{\text{bdy}} \lambda_{\text{lose}}$ (see (5.4)), which depends on $I, J$; for simplicity we ignore this case in what follows. The overall probabilities of each move type are chosen as
\[ \Lambda_{\text{gain}}(x) = \lambda_{\text{gain}} 1_{n_{\text{gain}} > 0}(x), \quad \Lambda_{\text{lose}}(x) = \lambda_{\text{lose}} 1_{n_{\text{lose}} > 0}(x), \quad \Lambda_{\text{same}}(x) = 1 - \Lambda_{\text{gain}}(x) - \Lambda_{\text{lose}}(x). \]
(3.19)
The difference from (3.9) is the specific choice of $\lambda_{\text{gain}}$, and the boundary cutoff $\sigma_{\text{bdy}}$.

We choose the move type using the probabilities in (3.19). If we propose a Gain or Lose move then we choose the labels uniformly from the neighbour sets:
\[ \text{Gain: } J \sim \text{Unif}(N_{\text{gain}}(I)), \quad \text{Lose: } J \sim \text{Unif}(N_{\text{lose}}^\text{bdy}(I, x)). \]
(3.20)
The densities for the label proposals are
\[ \text{Same: } \lambda_{I J}(x) = \Lambda_{\text{same}}(x), \quad \text{Gain: } \lambda_{I J}(x) = \frac{\Lambda_{\text{gain}}(x)}{n_{\text{gain}}(I)}, \quad \text{Lose: } \lambda_{I J}(x) = \frac{\Lambda_{\text{lose}}(x)}{n_{\text{lose}}(I, x)}. \]
(3.21)

Same move We use the same method and density introduced earlier, in (3.11) and the surrounding discussion. The overall density of proposing a Same move is
\[ \lambda_{I J}(x) h_{I J}(y|x) = \Lambda_{\text{same}}(x) \frac{1}{\sqrt{(2\pi\sigma^2)^d}} e^{-\frac{\|y-x\|^2}{2\sigma^2}} |T_{x,I}^T T_{y,I}| \Lambda_{\text{same}}(x) (y). \]
(3.22)
Note that in practice we do not have to calculate the Jacobian factor $|T_{x,I}^T T_{y,I}|$ for a Same move, since this factor cancels out in the acceptance ratio (3.1).

Gain move Consider a Gain move from $I \to J$ starting at $x \in M_I$, where $q$ is the dropped constraint. We say the move is one-sided if $q \in J_{\text{in}}$, so it becomes an inequality $q(x) > 0$, and we say the move is two-sided if $q \notin J_{\text{in}}$, so the constraint is simply forgotten. Given $x \in M_I$, a unit vector $u_n \in T_{x,J}$ that is normal to $T_{x,J}$, i.e. normal to the “boundary” $M_I$, is
\[ u_n = \frac{P_{x,J} \nabla q(x)}{|P_{x,J} \nabla q(x)|}, \quad \text{where } P_{x,J} = T_{x,J} T_{x,J}^T \]
(3.23)
is the orthogonal projection matrix onto $T_{x,J}$. Recall (3.14), where a similar vector was defined to construct Lose neighbours.

The tangent step $V \in T_{x,J}$ for a Gain move is constructed in two steps. First, we choose the component of the step that is normal to the boundary, in the direction of $u_n$. This component, call it $V_n = V \cdot u_n \in \mathbb{R}$,
has unit length in the normal direction and then projecting onto the tangent space:

The choice for a one-sided move ensures it always moves directly away from the boundary.

Next, given the value of \( V_n \), we choose a step in the tangential directions as an isotropic Gaussian with standard deviation \( \sigma_{\text{tan}}|V_n| \), where \( \sigma_{\text{tan}} > 0 \) is another parameter. The vector \( V_t \in \mathbb{R}^{d-1} \) representing the tangent components is generated as

\[
(V_t)_i \sim N(0, \sigma_{\text{tan}}^2 V^2), \quad 1 = 1, \ldots, d_j - 1.
\]

The proposal step is constructed from its normal and tangential components, observing that \( T_{x,J} = T_{x,J,u} \), as

\[
V = u_n V_n + T_{x,J} V_t.
\]

For small \( \sigma_{\text{tan}} \), the proposal mostly moves away from the boundary.

The density for this proposal is

\[
p^\perp(t V; x) = p^\perp g^\perp(V; V_n) p^\perp g^\perp(v_t | V_n)
\]

where the densities for the components of the step normal to (in the one-sided case), and tangential to the boundary, are, respectively,

\[
p^\perp g^\perp(V; V_n) = \frac{1}{\sigma_{\text{bdy}}} 1_{[0, \sigma_{\text{bdy}}]}(V_n), \quad p^\perp g^\perp(v_t | V_n) = \frac{1}{\sqrt{(2\pi\sigma_{\text{tan}}^2 V^2)^d}} e^{-\frac{|v_t|^2}{2\sigma_{\text{tan}}^2 V^2}}.
\]

For the two-sided case, the normal density is \( p^\perp g^\perp(V; V_n) = \frac{1}{2\sigma_{\text{bdy}}} 1_{[-\sigma_{\text{bdy}}, \sigma_{\text{bdy}}]}(V_n) \). We have omitted the densities’ dependence on \( I, J \) in the notation.

Putting the proposal densities (3.6), (3.27) together, the overall probability density with respect to \( \mu_J(dx) \) of proposing a one-sided Gain move \( (x, I) \rightarrow (y, J) \) is

\[
\lambda_{I,J}(x) h_{I,J}(y | x) = \frac{A_{\text{gain}}(x)}{n_{\text{gain}}} \frac{1}{\sigma_{\text{bdy}}} 1_{[0, \sigma_{\text{bdy}}]}(V_n) \frac{1}{\sqrt{(2\pi\sigma_{\text{tan}}^2 V^2)^d}} e^{-\frac{|v_t|^2}{2\sigma_{\text{tan}}^2 V^2}} |T_{x,J}^T T_{y,J}| A_{\text{gain}}(x)(y).
\]

For a two-sided move the modification is straightforward.

**Lose move** The direction \( V \) for a Lose move from \( I \rightarrow J \) at \( x \in M_I \) where \( q \) is the additional constraint is constructed as follows. Let \( v_{\text{opt}} = v_{\text{opt}}(x, I, q) \) as in (3.14) be the direction estimated to give the shortest distance to the boundary. We choose \( V \) by choosing a deterministic component in the direction of \( v_{\text{opt}} \), and random isotropic components \( R \in \mathbb{R}^{d-1} \) in the directions orthogonal to \( v_{\text{opt}} \), and then normalizing:

\[
V = \frac{v_{\text{opt}} + T_{x,J,v_{\text{opt}}}}{|v_{\text{opt}} + T_{x,J,v_{\text{opt}}}|} R, \quad R_i \sim N(0, \sigma_{\text{tan}}^2), \quad i = 1, \ldots, d_j - 1.
\]

The parameter \( \sigma_{\text{tan}} \) is the same one used to construct the reversed, Gain move. If \( \sigma_{\text{tan}} \) is small, the proposal is mostly in the direction that is estimated to give the shortest distance to the boundary, hence, is mostly normal to the boundary.

Given a step \( v \), there is a unique \( r = r(v) \) used to construct the step. This \( r \) is found by scaling \( v \) so it has unit length in the normal direction and then projecting onto the tangent space:

\[
r(v) = \frac{T_{x,J,v_{\text{opt}}}^T v}{v \cdot v_{\text{opt}}} \quad \text{if } v \cdot v_{\text{opt}} > 0.
\]
If \( v \cdot v_{\text{opt}} < 0 \), then \( r(v) \) is undefined; such a step could never be proposed.

Because we can solve uniquely for \( r(v) \), and the density \( p_R(r) \) for \( R \) is known analytically, we can obtain an analytic expression for the density in \( v \), as \( p_{IJ}^{\text{lose}}(v;x) = p_R(r) \frac{\partial r}{\partial v} |_{r, v_{\text{opt}} > 0} \). Here \( \frac{\partial r}{\partial v} \) is the determinant of the Jacobian of the transformation \( v \to r \). Since there are no constraints on \( r \), the Jacobian of the reverse transformation may be directly calculated from (3.30) to be

\[
\frac{\partial v}{\partial r} = \frac{(I - vv^T)T_{x,I,v_{\text{opt}}}}{|v_{\text{opt}} + T_{x,I,v_{\text{opt}}} r(v)|}.
\]

Substituting for \( r \) in the denominator using (3.31) shows that \( |v_{\text{opt}} + T_{x,I,v_{\text{opt}}} r(v)| = |v / v_{\text{opt}}| = |v_{\text{opt}}|^{-1} \).

For the numerator, we claim that its pseudodeterminant is

\[
|I - vv^T|T_{x,I,v_{\text{opt}}} = |v_{\text{opt}}|.
\]

To see this, note that the left-hand side has the form \( |P_{AB}B| \), where \( P_{AB} \) is the orthogonal projector onto the subspace \( A = \text{span}\{v\}^\perp \), and \( B = T_{x,I,v_{\text{opt}}} \) is a matrix with orthonormal columns spanning subspace \( B = T_{x,I,v_{\text{opt}}} \). By Theorem 1 in [5] and the subsequent discussion, \( |P_{AB}B| = \prod_k \cos \theta_k \), the product of the cosines of the principal angles \( \{\theta_k\}_k \) between subspaces \( A, B \). But the nonzero principal angles between \( A, B \) equal the nonzero principal angles between their complements, \( A^\perp, B^\perp \) [28, 30]. Since \( A^\perp = \text{span}\{v\} \subset B \), the largest principal angle between \( A^\perp, B^\perp \) is the angle between \( v, v_{\text{opt}} \), and the other principal angles are 0. This shows (3.33).

Therefore

\[
\left| \frac{\partial r}{\partial v} \right| = |v \cdot v_{\text{opt}}|^{-d_I},
\]

so the tangent step density is

\[
p_{IJ}^{\text{lose}}(v;x) = \frac{1}{\sqrt{(2\pi \sigma_{\text{tan}}^2)^d_I}} e^{-\frac{|v|^2}{2\sigma_{\text{tan}}^2}} |v \cdot v_{\text{opt}}|^{-d_I} 1_{v \cdot v_{\text{opt}} > 0}(v).
\]

Putting the proposal densities (3.8), (3.35) together, the overall probability density (with respect to \( \mu_I(dx) \)) of proposing a Lose move \((x,I) \to (y,J)\) is

\[
\lambda_{IJ}(x) h_{IJ}(y|x) = \frac{\Lambda_{\text{lose}}(x)}{n_{\text{lose}}} \frac{1}{\sqrt{(2\pi \sigma_{\text{tan}}^2)^d_J}} e^{-\frac{|y|^2}{2\sigma_{\text{tan}}^2}} |v \cdot v_{\text{opt}}|^{-d_J} 1_{v \cdot v_{\text{opt}} > 0}(v) T_{x,I,J} T_{y,J} |y - x|^{-d_J} 1_{A_{\text{loss}}(x)}(y).
\]

### 4 Examples

We now return to our examples, and illustrate the type of information that can be gained by sampling probability distribution on each manifold (Section 4.1). We also show that our sampler gives a good approximation to the probability distribution for a real system with an interaction potential that does not have infinitesimal range. We then discuss the efficiency of the method (Section 4.2), and clarify for which systems it is expected to give a significant computational speedup. The codes used to run all of the following examples are available at [11].

#### 4.1 Revisited examples

We revisit in turn each example from Section 2.3.
### 4.1 Example [1] Parabola and line in two dimensions

We found Example [1] to be useful when developing our code, as it contains most of the special cases one must handle (e.g. manifolds with no constraints, zero-dimensional manifolds, manifolds with no Lose or Gain neighbours, manifolds with both Lose and Gain neighbours, flat manifolds, curved manifolds), yet it can be visualized easily and many statistics are known analytically.

We sampled the probability measure $\rho(dx)$ defined in (2.15), which gives equal weight to the arclength measures on each manifold. Figure 9 shows that the total fraction of time spent on each manifold, as well the marginal distributions in $x$ on each manifold, agree very well with the analytically calculated distributions.

### 4.1.2 Example [2] a trimer of discs in two dimensions

Consider configurations of three two-dimensional discs with radius $r = 0.5$, where all bonds have the same sticky parameter $\kappa$. The stratification is $S = M_I \cup M_J$ where $I$ is the Polymer and $J$ is the Triangle. We sampled the probability measure $\rho(x)dx$ defined in (2.12), which takes the specific form

$$
\rho_{trimer}(dx) = Z^{-1} \left( \kappa^2 |Q_I(x)|^{-1} \mu_I(dx) + \kappa^3 |Q_J(x)|^{-1} \mu_J(dx) \right).
$$

(4.1)

Here $Z$ is the normalizing constant. The probability of finding the cluster in the Polymer or Triangle is found by integrating $\rho_{trimer}(dx)$ over the space of rotations and internal motions for each kind of cluster, which can be done analytically for this simple example. The Triangle has no internal motions, so its probability is

$$
P_{Triangle} \frac{2\pi Z^{-1}}{2\pi Z^{-1}} = \kappa^3 2 |Q_J(x)|^{-1} |x_{cm}| = \frac{4\sqrt{3}}{9} \kappa^3.
$$

Here $x_{cm}$ is obtained from a configuration $x$ of the triangle by translating it so its center of mass lies at the origin, the factor $2 |x_{cm}|$ comes from integrating over the cluster’s rotations, and the factor 2 comes from counting the two different copies of the triangle obtained by permuting particle labels. For the unit triangle, $|x_{cm}| = 1$, $|Q_J| = \sqrt{27}/4$.

The Polymer has one internal degree of freedom, which may be parameterized by its internal angle $\theta$. Following [21], we adopt the parameterization $x(\theta) = (-\sin \frac{\theta}{2}, -\frac{1}{3} \cos \frac{\theta}{2}, 0, \frac{2}{3} \cos \frac{\theta}{2}, \sin \frac{\theta}{2}, -\frac{1}{3} \cos \frac{\theta}{2})$, which keeps the center of mass of the cluster at the origin and doesn’t rotate it. The Hausdorff measure in this parameterization is $\mu_J(x(d\theta)) = \frac{1}{2\pi} d\theta$. The probability of the Polymer is

$$
P_{Polymer} \frac{2\pi Z^{-1}}{2\pi Z^{-1}} = \kappa^3 \int_{\theta=\pi/3}^{5\pi/3} |Q_I(x(\theta))|^{-1} |x(\theta)| \frac{dx}{d\theta} d\theta = \frac{4\pi}{9} \kappa^2.
$$
To evaluate the integral we calculated \(|Q_I(x(\theta))| = \sqrt{4 - \cos^2 \theta}, |x(\theta)| = \sqrt{\frac{2}{3} + \frac{4}{3} \sin^2 \frac{\theta}{2}}, \int |dx| = \frac{1}{2} \sqrt{\frac{2}{3} + \frac{4}{3} \cos^2 \frac{\theta}{2}};\) the integrand therefore is \(\frac{1}{2}\).

Putting these calculations together shows the probability of the Triangle is \(P_{\text{Triangle}} = \frac{\kappa}{\kappa + \pi/\sqrt{3}}.\) (4.2)

This probability is plotting in Figure 10 along with estimates obtained from sampling \(\rho_{\text{trimer}}(dx)\) in (4.1) using our algorithm. This figure also shows that the sampled probability distribution of the internal angle \(\theta\) conditional on being in the Polymer state, estimated from a sampling run with \(10^7\) points and \(\kappa = 1\) and other parameters as above. The distribution is flat, as expected. Right: Rejection rates for these sampling runs. For Gain moves (Triangle \(\rightarrow\) Polymer) and Lose moves (Polymer \(\rightarrow\) Triangle) all rejections came from the Metropolis step. For Same moves, the rejection rates were uniformly 0.05 on the Triangle (all from the NES failing), and 0.44 on the Polymer (0.25 from NES; 0.12 from Metropolis step; 0.07 from violating inequalities.)

To evaluate the integral we calculated \(|Q_I(x(\theta))| = \sqrt{4 - \cos^2 \theta}, |x(\theta)| = \sqrt{\frac{2}{3} + \frac{4}{3} \sin^2 \frac{\theta}{2}}, \int |dx| = \frac{1}{2} \sqrt{\frac{2}{3} + \frac{4}{3} \cos^2 \frac{\theta}{2}};\) the integrand therefore is \(\frac{1}{2}\).

Putting these calculations together shows the probability of the Triangle is \(P_{\text{Triangle}} = \frac{\kappa}{\kappa + \pi/\sqrt{3}}.\) (4.2)

This probability is plotting in Figure 10 along with estimates obtained from sampling \(\rho_{\text{trimer}}(dx)\) in (4.1) using our algorithm. This figure also shows that the sampled probability distribution of the internal angle \(\theta\) conditional on being in the Polymer state, estimated from a sampling run with \(10^7\) points and \(\kappa = 1\) and other parameters as above. The distribution is flat, as expected. Right: Rejection rates for these sampling runs. For Gain moves (Triangle \(\rightarrow\) Polymer) and Lose moves (Polymer \(\rightarrow\) Triangle) all rejections came from the Metropolis step. For Same moves, the rejection rates were uniformly 0.05 on the Triangle (all from the NES failing), and 0.44 on the Polymer (0.25 from NES; 0.12 from Metropolis step; 0.07 from violating inequalities.)

4.1.3 Example 3: a polymer of 6 sticky unit spheres in three dimensions

Identical Interactions We first treated all interactions as identical, and sampled the measure \(\rho_{\text{equal}}\) in (2.16) with sticky parameter \(\kappa_0 = 2\) for all bonds, recording the number of bonds at each time step. From this data we can recover the probability \(p_m\) of observing \(m\) bonds at any sticky parameter \(\kappa\), by reweighting the data as follows. Integrating \(\rho_{\text{equal}}\) over manifolds with \(m\) bonds, gives that

\[
p_m(\kappa) = \frac{\kappa^m Z_m}{\sum_{m=5}^{12} \kappa^m Z_m}, \quad Z_m = \sum_{l:|l|=m} \int_{\mu_l} |Q_j(x)|^{-1} \mu(x) dx.
\]

The quantity \(\kappa^m Z_m\) is the partition function for the set of configurations with \(m\) bonds. We call \(Z_m\) the geometrical partition function because it depends only on the geometry of the spheres, but not on the strengths of their interactions or the temperature; it is a measure of the entropy of the \(m\)-bonded configurations.

We can estimate the ratio of geometrical partition functions from our data, as

\[
\frac{Z_m}{Z_{m-1}} \approx \frac{1}{\kappa_0} \frac{T_m}{T_{m-1}},
\]

where \(T_m\) is the time the simulation spent in the states with \(m\) bonds. Up to a normalizing constant, an estimate \(\hat{Z}_m\) of the geometric partition functions is found by setting \(\hat{Z}_5 = 1\), and then recursively calculating \(\hat{Z}_m = \kappa_0^{-1} \hat{Z}_{m-1} T_m / T_{m-1}\). From these estimates we computed the probabilities \(p_m(\kappa)\) at any value of \(\kappa\) using (4.3). These probabilities are shown for a range of \(\kappa\) in Figure 11.
Figure 11: For Example 3, the probability of finding a polymer of 6 unit spheres with each given number of bonds as a function of the sticky parameter $\kappa$, the strength of the interactions between off-backbone spheres. Solid lines show the probabilities inferred from the Stratification Sampler at $\kappa = 2$ with $4 \times 10^7$ points (recording data every 4 points), and sampling parameters $\sigma = 0.4$, $\sigma_{bdy} = 0.3$, $\sigma_{\tan} = 0.2$, $\lambda_{\text{lose}} = 0.4$, $\lambda_{\text{gain}} = 0.24$. Markers show the probabilities estimated from Brownian dynamics simulations run to simulation time $10^4$ at different values of $\kappa$, using a Morse potential for the off-backbone interactions and a spring potential for the backbone interactions as in (4.5), (4.6). The agreement between the statistics from the Stratification sampler and Brownian dynamics is excellent, verifying both that statistics from a simple run of the Stratification sampler can be extrapolated to other parameter values, and that the Stratification sampler gives good predictions even for a system whose interactions have small but nonzero range.

Comparing with Brownian Dynamics simulations

We wish to verify two things: one, that these reweighted probabilities are correct, and two, that our algorithm is a good model for a real system of particles, which is never exactly at the sticky limit. To this aim we perform Brownian dynamics simulations of particles with short-ranged, strong interaction potentials, which the sticky limit is intended to model. We construct an energy $U(x)$ as

$$U(x) = \sum_{(i,j) \in E: j = i+1} U_{ij}^{\text{spring}}(|x_i - x_j|) + \sum_{(i,j) \in E: j \neq i+1} U_{ij}^{\text{morse}}(|x_i - x_j|).$$  \hspace{1cm} (4.5)

Here

$$U_{ij}^{\text{spring}}(r) = \frac{1}{2} k_{\text{spring}} (r - d_{ij})^2, \quad U_{ij}^{\text{morse}}(r) = E (1 - e^{-\rho (r-d_{ij})})^2 - E$$

are a spring potential, and Morse potential, respectively. The quantity $d_{ij}$ is the distance where spheres are exactly touching; in our example $d_{ij} = 1$. The spring potential keeps the bonds in the backbone fixed, and the parameter $k_{\text{spring}}$ controls how much particles typically deviate from perfect contact; it is roughly the square of the inverse width of the interaction. The Morse potential creates a strong bond between non-backbone pairs when the distance between their surfaces is less than $\approx 2.5/\rho$, and is otherwise nearly zero so particles don’t feel each other. We used a spring potential for the backbone as an alternative to a Morse potential with very large $E$, as the latter would require prohibitively small timesteps to resolve numerically.

We numerically simulated the system

$$dX_t = -\nabla U(X_t) dt + \sqrt{2} dW_t, \hspace{1cm} (4.6)$$

where $X_t \in \mathbb{R}^{6 \cdot 3}$ is the configuration of the system and $W_t \in \mathbb{R}^{6 \cdot 3}$ is a Brownian motion, using an Euler-Maruyama method with time step $\Delta t$\footnote{The Euler-Maruyama method constructs a numerical approximation $X_0, X_1, X_2, \ldots$ to the solution to (4.6) at times 0, $\Delta t, 2\Delta t, \ldots$}. The stationary distribution for (4.6) when constrained to a box $\mathbb{B}$
is $Z^{-1}e^{-U(x)}$ with $Z = \int e^{-U(x)}$; this approaches $\rho_{\text{equal}}$ as $k_{\text{spring}}, \rho \to \infty$ and $E \to \infty$. We chose width parameters $\rho = 60$, $k_{\text{spring}} = 6\rho^2$ which gave us roughly the same width for the spring potential as for the Morse potential, and varied $E$. The choice of range is characteristic of certain DNA-mediated interactions (though some have smaller range \cite{55}); it was small enough that the sticky limit gives a good description of the set of states on the energy landscape \cite{52}, but large enough that it was not too prohibitive to resolve numerically. We needed a timestep of $\Delta t = 10^{-6}$ to resolve the interactions.

We recorded the number of bonds in configuration $X$, every 0.05 units of time. We said a pair $(i, j)$ was bonded if $|x_i - x_j| < 1 + 2.5/\rho$ \cite{52}. We verified that changing the cutoff (from about $2/\rho$ to $4/\rho$) did not significantly change the measured statistics.

To compare to the Stratification sampler we determined the effective sticky parameter at each value of $E$ as

$$\kappa(E) = \int_0^{1 + 2.5/\rho} e^{-U(r; E)} dr. \quad (4.7)$$

We calculated the integral by numerical integration, as Laplace asymptotics were not accurate enough for small values of $E$.

The empirical probabilities $p_m(\kappa)$ estimated from the Brownian dynamics simulations at several values of $\kappa$ are shown in Figure 11. These agree with the probabilities predicted from the Stratification sampler. This not only verifies the correctness of the sampler, but it also shows that the sampler gives a good approximation to a real system with a non-delta-function potential.

**Non-identical interactions: Designing interactions for self-assembly**

Next we explain how to address inverse problems in self-assembly, where we wish to choose non-identical interactions to make the probability of observing a desired target cluster as high as possible. Consider the lowest-energy clusters for the system, which are the clusters with $m = 12$ bonds; we call these clusters “rigid” since they are mechanically rigid when represented as a graph with edges between spheres (vertices) in contact. There are two distinct rigid clusters, after lumping together clusters with isomorphic adjacency matrices – an octahedron and a polytetrahedron, illustrated in Figure 4. These occurred with relative probabilities 5%, 95% respectively. It was shown in \cite{26} that the octahedron occurs much less frequently because it has many more symmetries in its point group.

Suppose we wish to form an octahedron in equilibrium with high probability. This is impossible with identical particles, since changing the sticky parameter only changes the probabilities $p_m$ of having $m$ bonds in some cluster, but not the relative probabilities of finding each particular cluster with $m$ bonds. Therefore, we suppose the particles are one of two types, labelled “A” and “B” as in Figure 4, with sticky parameters $\kappa_{AA}, \kappa_{AB}, \kappa_{BB}$ for interactions between the given types, so the equilibrium distribution is \cite{24}. We reweighted our data from a sampling run with identical sticky parameter $\kappa_0 = 2$, using weights for each sampled point $X_k, I_k$ constructed as

$$w(X_k, I_k) = \left(\frac{\kappa_{AA}}{\kappa_0}\right)^{n_{aa}(I_k)} \left(\frac{\kappa_{AB}}{\kappa_0}\right)^{n_{ab}(I_k)} \left(\frac{\kappa_{BB}}{\kappa_0}\right)^{n_{bb}(I_k)}.$$

From the reweighted data we calculated the relative probability of the octahedron, for each choice of parameters $\kappa_{AA}, \kappa_{AB}, \kappa_{BB}$.

Figure 12 shows that when $\kappa_{BB} = 0$, the octahedron forms with 100% probability as $\kappa_{AA} \to 0$ and $\kappa_{AB} \to \infty$. However, the probability of observing a rigid cluster goes to 0 in this limit, so the overall probability of observing the octahedron also goes to zero for these parameter choices. This observation is consistent with \cite{26}, which used a mean-field approach to argue that the best assembly occurs when

$$X_{k+1} = X_k + -\nabla U(X_k)\Delta t + \sqrt{2\Delta t} \xi_k,$$

where $\xi_1, \xi_2, \ldots \sim N(0, 1)$ is a sequence of independent standard normal random variables.

The numbers in our data are slightly different from those in Meng et al \cite{39}, because we consider the backbone to be fixed, while they allow all bonds to break and form, which changes the symmetry number for each cluster.
Figure 12: Studying inverse problems in interaction design, for $N = 6$ unit spheres with two types of particles A, B as shown in Figure 4 and explained in Example 3. Left column: equilibrium probability of observing an octahedron, given that the system is a rigid cluster (a cluster with $m = 12$ bonds), for varying sticky parameters $\kappa_{AA}, \kappa_{AB}$, and for $\kappa_{BB} = 0, 0.1$. Right column: equilibrium probability of observing a rigid cluster as a function of $\kappa_{AA}, \kappa_{AB}$, for $\kappa_{BB} = 0, 0.1$. The top row shows the octahedron forms much more often than the polytetrahedron with small $\kappa_{AA}, \kappa_{AB}$ and large $\kappa_{BB}$ (top left), however the probability of forming a rigid cluster is small in this limit (top right). The bottom row shows the octahedron form with high probability when $\kappa_{BB}$ is small, $\kappa_{AB} \sim O(1)$, and $\kappa_{AA}$ is large (bottom left), and furthermore, the probability of forming a rigid cluster is reasonably high in this limit (bottom right.)
Figure 13: For Example 4, the average fraction of particles on the wall (left) and average radius of gyration $R_g = |x_1 - x_N|$ (middle) as a function of wall sticky parameter $\kappa$, for several values of $N$ as indicated in the legends, and for a semiflexible polymer with $N = 20$ and $\kappa_{\text{bend}} = 2$ (see text.) For the semiflexible polymer we plot $R_g/2$ instead of $R_g$, to ease comparison. Markers indicate the values of $\kappa$ at which simulations were run, and curves indicate the averages estimated by reweighting data from nearby markers, in a similar way to (4.8). Each estimate was formed from $10^7$ simulation steps saved every 10 steps, with parameters $\sigma_{\text{bdy}} = 0.3, \sigma_{\text{tan}} = 0.2, \lambda_{\text{lose}} = 0.4, \lambda_{\text{gain}} = 2\sigma_{\text{bdy}} \lambda_{\text{lose}}$, for the freely jointed polymer we used $\sigma = 0.3, 0.2, 0.15, 0.12$ for $N = 10, 20, 40, 80$ respectively and for the semiflexible polymer we used $\sigma = 0.15$. The right plot shows the time in minutes taken by each simulation with $\kappa = 1$ on a 2.40GHz Intel Xeon CPU E5-2680; on a log scale, the data is well-fit by a line with slope 2.5.

sticky parameters are either zero or the same constant value; there are no intermediate maxima for the probability.

In practice, it is hard to create sticky parameters that are exactly zero, especially for DNA-coated colloids where there is always some weak binding between non-complementary DNA strands \[27\]. Therefore we also consider $\kappa_{\text{BB}} = 0.1$, and find the octahedron forms with 100% probability for $\kappa_{\text{AA}} = O(1)$, and $\kappa_{\text{AB}} \to \infty$. In this limit the probability of observing a rigid cluster also goes to 100%. Therefore, it is possible to form the octahedron with high probability, with the right choice of parameters.

A tentative explanation for what makes the octahedron have high probability is that with 2 Bs, the polymer can be folded into an octahedron such that each B particle is in contact with 4 A particles, but neither B particle is in contact with itself. On the other hand, if the polymer folds into the polytetrahedron, then if the 2 Bs are each in contact with 4 As, then they must also be in contact with each other, forming a weak bond that effectively wastes a precious A-A contact. We explored other labellings of the particles, and tentatively found that with any ordering of 2 nonadjacent Bs and 4 As, the octahedron formed with high probability under the same conditions, however with 3 Bs and 3 As we could not form the octahedron with high probability. It would be interesting to explore these observations further, and to find a general principle for the how the particle types and locations should be chosen for a polymer of $N$ spheres to fold into a given cluster.

4.1.4 Example 4 Polymer adsorbing to a flat surface

We sampled the distribution (2.18) for a freely-jointed polymer for several values of $N$, and the distribution (2.19) for a semi-flexible polymer for $N = 20$, each at seven different values of sticky parameter, $\kappa \in \mathcal{K} = \{5^{j/2}\}_{j=3}^3$. For each $N$, we computed two statistics as a function of $\kappa$, the average fraction of spheres on the surface $f$, and the average radius of gyration, $R_g = |x_1 - x_N|$, see Figure 13. For values of $\kappa$ not in the list $\mathcal{X}$, we estimated these averages by reweighting the data at the values in $\mathcal{X}$, constructing weights in a similar manner to (4.8) and interpolating the estimates from each value in $\mathcal{X}$. We found that, with the exception of $N \leq 10$, reweighting the data from a single simulation could not adequately capture the statistics over the full range of $\kappa$ values. This is because as $N$ increases, the distributions for each statistic become increasingly concentrated near their average values, so there is inadequate data at other values of the statistic to extrapolate far from the mean. However, for the values in $\mathcal{K}$, there is still enough overlap
in the reweighted distributions to make extrapolation and interpolation meaningful.

As expected, \( f \) increases with \( \kappa \), from \( \approx 0 \) at small \( \kappa \), when no particles are on the surface, to 1 at large \( \kappa \), where all particles are on the surface, with a sharp transition region whose width and location appear relatively independent of \( N \), though which is slightly sharper for the semiflexible polymer. The radius of gyration has interesting non-monotonic behaviour: for a freely jointed polymer, it appears to be slightly larger than \( \sqrt{N-1} \) for small \( \kappa \), and slightly smaller than \( \sqrt{N-1} \) for larger \( \kappa \), with a dip in between. That it is not always \( \sqrt{N-1} \), its theoretical value for a polymer in free space of any dimension, must be due to the boundary condition at the surface. The radius of gyration for a semiflexible polymer has the opposite behaviour, increasing with \( \kappa \), but also nonmonotonically; overall it is nearly twice as large as for a freely jointed polymer.

We remark that our observations are not new, but we present them here to show the variety of systems or constraints that can be studied by our sampler. An easy modification that could give new information would be to change the surface to which the polymer is adsorbing to one that is curved.

4.1.5 Example 5 Surface area of a 10-dimensional ellipsoid

We sampled the distribution \((2.21)\) for \( n = 10 \), using semiaxes \( (a_1, \ldots, a_{10}) = (2, 2, 2, 3, 3, 3, 3, 1, 1, 1) \) and choosing the weights to be \( c_k = e^{0.94k} \). This choice was motivated by an initial sampling run with equal weights in which the probability to be in \( I^k \) was approximately proportional to \( e^{-0.94k} \). We used sampling parameters \( \sigma = 0.6, \sigma_{\text{tan}} = 0.3, \sigma_{\text{bdy}} = 0.4, \lambda_{\text{lose}} = 0.4, \lambda_{\text{spin}} = \sigma_{\text{bdy}} \lambda_{\text{lose}} = 0.16 \). We ran the sampler for \( 10^7 \) steps and used \((2.22)\) to estimate the surface area, dividing the data into 10 bins to estimate a one standard deviation error bar, and obtained

\[
\text{Vol}(E_{10}) \approx 7155 \pm 162,
\]

about a 2% relative error.

To check the accuracy of this estimate, we computed it in a different way, by constructing an alternative stratification using \( J = \{(q_1), \emptyset\} \cup (\emptyset, \{-q_1\}) \). This stratification consists of the surface of \( E_{10} \) and its interior. The volume of the interior is known analytically to be \( a_1 a_2 \cdots a_{10} \cdot \pi^5 / 120 \), so an estimate for the surface area is

\[
\text{Vol}(E_{10}) \approx \frac{\# \text{ of points in } E_n}{\# \text{ of points in interior}(E_n)} \cdot \frac{\pi^5 \cdot a_1 a_2 \cdots a_{10}}{120} = 7138 \pm 21,
\]

The relative error is about 0.2%, with the same number of points as for the previous stratification. For this estimate we used the same sampling parameters as for the first estimate. While the second stratification clearly gives a more accurate estimate, because it is formed from fewer manifolds, one does not usually know the volume of a high-dimensional shape analytically, so this simpler method cannot usually be applied.

We remark that this strategy is a form of thermodynamic integration, but where \emph{dimension} is the variable that changes, rather than temperature [15]. For higher-dimensional volumes, where the sampler spends most of the time in the intermediate dimensions of the stratification, an alternative to choosing non-equal weights \( c_k \) would be to break up the problem into several separate sampling problems: first estimate the ratio of the \( d \)-dimensional volumes in the stratification to the \( d-1 \)-dimensional volumes, then the ratio of \( d-2 \)-dimensional to \( d-3 \)-dimensional, and so on down to the ratio of one-dimensional to 0-dimensional volumes. For a variant of this strategy applied to shrinking sets, not changing dimensions, see [30] [50].

4.2 Efficiency of the Stratification sampler

We use our examples to empirically investigate the efficiency of the Stratification sampler.

First, consider Example 3 a polymer of 6 unit spheres in three dimensions. We obtained the same statistics from the Stratification sampler, as from a more conventional Brownian dynamics simulation. Which method is more efficient? We ran both methods with sticky parameter \( \kappa = 2.885 \), and computed
the estimates $\hat{m}_S^m$, $\hat{m}_S^{BD}$ for $p_m(2.885)$. We estimated the standard deviations $\sigma_m^S$, $\sigma_m^{BD}$ of each estimate by binning the data into 8 bins. The Stratification sampler was run for $10^7$ steps and the Brownian dynamics simulation was run for $10^7$ simulation time units. Table 1 shows that under these conditions, the standard deviations (a measure of error) for the Stratification sampler are about 2.5–7 times smaller than those for Brownian dynamics.

The standard deviation is not a good measure of efficiency, since the Stratification Sampler took much less physical time to run: it took 815 seconds (just under 14 minutes), compared to 17900 seconds (just under 5 hours) for Brownian Dynamics on the same processor – about 22 times less time. Assuming that the simulation time scales with the inverse of the standard deviation squared, as for a typical Monte Carlo simulation, we can estimate the ratio of the physical times it would take each method to run to achieve the same value of standard deviation as $(\sigma_m^{BD}/\sigma_m^S)^2 \cdot (17900/815)$. These predicted ratios of physical times are shown in the last column of Table 1, they range from about 150 to over 1000. That is, the Stratification sampler is two to three orders of magnitude faster than Brownian dynamics for this example, to achieve a given level of accuracy.

Next we consider how the efficiency of the Stratification sampler scales with the size of the system. For this we return to Example 4, a polymer weakly adsorbing to a flat surface. Figure 13 also compares the amount of time it took to run a simulation with each of $N = 10, 20, 40, 80$ particles for $10^7$ steps with $\kappa = 1$. The time increases rapidly with $N$: it is well-approximated by a power law $\propto N^{2.5}$. This is worse than the $O(N)$ increase in computation required by Brownian dynamics to simulate a polymer with no excluded volume interactions (no overlaps), and also worse than the $O(N \log N)$ increase with excluded volume interactions [16].

We have implemented our algorithm in the most naive way possible, using dense linear algebra to find the tangent space, via the QR decomposition, and to project back to the manifold using Newton’s method, applying the LU decomposition to solve systems of nonlinear equations. Our choices lead to poor scalings with system size: for very large $N$, we expect the computational time to scale as $O(N^3)$, since this is the scaling of both the QR-factorization and the LU-decomposition [14, 51]. These linear algebra calculations can be made more efficient for problems with special structures, as we discuss in the conclusion (Section 6); our naive implementation should be used only for systems with no more than a few dozen variables, where it does give a large speedup despite the poorer scaling with system size.

Table 1: Estimates of $p_m$ computed using the Stratification sampler (same parameters as in Figure 11) and by Brownian dynamics, both with $\kappa = 2.885$. The Stratification sampler ran for $10^7$ steps and took a physical time of 13.6 minutes on a 2.40GHz Intel Xeon CPU E5-2680. The Brownian dynamics simulation ran for $10^7$ simulation time units and took a physical time of 5 hours on the same computer. The standard deviations $\sigma_m^S$, $\sigma_m^{BD}$ for each estimate $\hat{m}_m$ were calculated by binning the data into 8 bins; the standard deviation gives a measure of the error of the estimate. The ratio of errors $\sigma_m^{BD}/\sigma_m^S$ is shown in the second-last column, however it is a misleading comparison since the simulations took different amounts of physical time. By multiplying the squared ratio of errors by the ratio of physical simulation times, we obtain the ratio of physical simulation times that would be required for each method to produce the same given error, shown in the last column.

| # bonds | Stratification sampler $\hat{m}_m^S \pm \sigma_m^S$ | Brownian Dynamics $\hat{m}_m^{BD} \pm \sigma_m^{BD}$ | Ratio of Errors $\sigma_m^{BD}/\sigma_m^S$ | Ratio of Physical Times for fixed error |
|---------|--------------------------------------------------|--------------------------------------------------|------------------------------------------|--------------------------------------|
| 12      | 0.1541 ± 0.00054                                 | 0.1568 ± 0.00147                                 | 2.7                                      | 162                                  |
| 11      | 0.2675 ± 0.00048                                 | 0.2651 ± 0.00259                                 | 5.4                                      | 640                                  |
| 10      | 0.2598 ± 0.00021                                 | 0.2556 ± 0.00152                                 | 7.2                                      | 1155                                 |
| 9       | 0.1799 ± 0.00046                                 | 0.1798 ± 0.00184                                 | 4.0                                      | 358                                  |
| 8       | 0.0916 ± 0.00039                                 | 0.0919 ± 0.00146                                 | 3.7                                      | 304                                  |
| 7       | 0.0352 ± 0.00028                                 | 0.0374 ± 0.00130                                 | 4.6                                      | 467                                  |
| 6       | 0.0102 ± 0.00015                                 | 0.0115 ± 0.00038                                 | 2.6                                      | 147                                  |
| 5       | 0.00177 ± 0.00006                                | 0.00197 ± 0.00017                                | 2.7                                      | 158                                  |
5 Acceptance probabilities for flat manifolds

This section shows that the choice of Gain and Lose proposal moves presented in Section 3.3.2, give 100% acceptance probabilities for a stratification consisting of two flat manifolds defined by affine constraints with no additional inequalities. We first motivate these proposals by considering in detail the simplest possible cases, a 0-dimensional point at the boundary of a 1-dimensional line, and then a 1-dimensional line at the boundary of a 2-dimensional plane. We explain how to construct proposals to obtain a 100% acceptance probability. This discussion also shows how one could choose other proposals and still retain 100% acceptance probability. Finally we show that the acceptance probability is 100% for the general linear case, a $d$-dimensional hyperplane forming the boundary for a $(d+1)$-dimensional hyperplane.

5.1 0-dimensional manifold ↔ 1-dimensional manifold

Let $x \in \mathbb{R}$, and define two manifolds $M_0$, $M_1$ by

$$M_0 = \{0\}, \quad M_1 = \{x \in \mathbb{R} : x > 0\}.$$  \hfill (5.1)

Let $S = M_0 \cup M_1$. One way to construct such a stratification via level sets is to define a function $q(x) = x$. Let $f_1(x), f_0$ be functions defining the probability measure on each manifold.

Let $\lambda_{\text{gain}}, \lambda_{\text{lose}}(x)$ be the probabilities of choosing a Gain and Lose move respectively from a given point (there is only one possible starting point for a Gain move, so we omit it in the notation.) Let the $v$-density for a Gain move be $p_{\text{gain}}(v)$, and let the $v$-density for a Lose move be $p_{\text{lose}}(v)$. Since there are only two unit vectors in the tangent plane to $M_1$, $\pm 1$, we make the decision that $p_{\text{lose}}(v) = \delta_{-1}(v)$, a unit mass at $v = -1$. This ensures we always propose to move in the direction of the boundary, not away from it. For a general one-dimensional affine constraint $q(x)$, we can determine which direction points toward the boundary by evaluating $dq/dx$.

Consider a Gain move from $x = 0$ to $y \in M_1$. The tangent step is $v = y$ and the reverse step is $v_{\text{rev}} = -y/|y| = -1$. The Jacobian factors are both 1 in this simple example, so the Metropolis ratio in the acceptance probability (3.1) is

$$a(y,1|x,0) = \frac{f_1(y)\lambda_{\text{lose}}h_{01}(x|y)}{f_0\lambda_{\text{gain}}h_{10}(y|x)} = \frac{f_1(y)}{f_0} \frac{\lambda_{\text{lose}}(y)}{\lambda_{\text{gain}}p_{\text{gain}}(y)}.$$  \hfill (5.2)

We wish to make the acceptance ratio as high as possible, to avoid wasting computation in proposing moves. For simplicity suppose that $f_1(y) = f_1$, a constant independent of $y$. For the acceptance probability to be 1, we need

$$\lambda_{\text{lose}}(y) = \frac{f_0}{f_1} \lambda_{\text{gain}}p_{\text{gain}}(y).$$  \hfill (5.3)

Since $p_{\text{gain}}(y)$ is a probability density, whose integral is 1, we also need that

$$\lambda_{\text{gain}} = \frac{f_1}{f_0} \int_0^\infty \lambda_{\text{lose}}(y) dy.$$  \hfill (5.4)
Figure 15: Setup for the example in Section 5.2, which constructs optimal proposal moves between the x-axis $M_1$ and the upper half plane $M_2$.

Therefore once we choose $p_{\text{gain}}(y)$, this sets the ratio $\lambda_{\text{lose}}(y)/\lambda_{\text{gain}}$ via (5.3), and conversely if we choose $\lambda_{\text{lose}}(y)$, this sets both $\lambda_{\text{gain}}$, via (5.4), and then $p_{\text{gain}}(y)$ via (5.3).

A proposal that can be implemented in practice is to choose a Lose move with constant probability within a certain cutoff distance $\sigma_{\text{bdy}}$ from the boundary. This implies that $p_{\text{gain}}(y)$ must be uniform on $[0, \sigma_{\text{bdy}}]$, so

$$\lambda_{\text{lose}}(y) = \lambda_{\text{lose}}I_{[0,\sigma_{\text{bdy}}]}(y), \quad p_{\text{gain}}(y) = \frac{1}{\sigma_{\text{bdy}}} I_{[0,\sigma_{\text{bdy}}]}(y), \quad \lambda_{\text{gain}} = \frac{f_1}{f_0} \sigma_{\text{bdy}} \lambda_{\text{lose}}. \quad (5.5)$$

There are two parameters defining these proposals, the cutoff $\sigma_{\text{bdy}}$, and the lose parameter $\lambda_{\text{lose}}$, which is subject to the constraint $0 < \lambda_{\text{lose}} < \min(1, \sigma_{\text{bdy}}^{-1})$. The probability of proposing a Same move along $M_1$ is then $\lambda_{\text{same}}(y) = 1$ if $y > \sigma_{\text{bdy}}$, and $\lambda_{\text{same}}(y) = 1 - \lambda_{\text{lose}}$ if $y < \sigma_{\text{bdy}}$. The probability of proposing a Same move on $M_0$ (repeating the point) is $\lambda_{\text{same}}(0) = 1 - \lambda_{\text{gain}}$.

5.2 1-dimensional manifold $\leftrightarrow$ 2-dimensional manifold

Next let $x \in \mathbb{R}$, and define two manifolds $M_1, M_2$ by

$$M_1 = \{(x_1, x_2) \in \mathbb{R}^2 : x_2 = 0\}, \quad M_2 = \{(x_1, x_2) \in \mathbb{R}^2 : x_2 > 0\}. \quad (5.6)$$

See Figure 15. For simplicity we assume that $f_1(x_1) = f_2(x_1, x_2) = \text{cst}$ everywhere. Let $e_1 = (1,0)$, $e_2 = (0,1)$. We may assume that proposal moves are homogeneous in the $x_1$ direction, so we must choose functions $\lambda_{\text{gain}}, \lambda_{\text{lose}}(x_2), p_{\text{gain}}(v), p_{\text{lose}}(\theta; x_2)$. Here $\theta$ is the direction of a proposed Lose move $v$, measured counterclockwise from $v_{\text{opt}} = -e_2$; the corresponding unit vector is $v = (\sin \theta, -\cos \theta)$.

Consider a Gain move from $x = (x_1, 0) \in M_1$ to $y = (y_1, y_2) \in M_2$. Since we assume homogeneity in $x_1$, we may assume without loss of generality that $x_1 = 0$. The forward tangent step is $v = y - x = (v_1, v_2) = (y_1, v_2)$. The Jacobian factor $|\frac{\partial v}{\partial y}|$ in (3.6) is $|T_{y_2}T_{y_2}| = 1$, since the step is along a plane.

The reverse tangent step is $v_{\text{rev}} = (v_{\text{rev},1}, v_{\text{rev},2}) = (y - x)/|y - x| = (\sin \theta, -\cos \theta)$ for some angle $\theta$. To calculate the jacobian factor $|\frac{\partial v}{\partial y}|$ in (3.8) for the Lose move from $y \rightarrow x$, we calculate $T_{y,2,v_{\text{rev}}} = (\cos \theta, \sin \theta)$, the unit vector perpendicular to $v_{\text{rev}}$. See Figure 15. We have that $T_{x,1} = e_1$, the tangent space to the horizontal axis. Therefore $|T_{x,1}| = |\text{cos} \theta|$. By geometry, $a = y_2/|\text{cos} \theta|$, and it is raised to the power of $d_j = 1$ (where $J$ represents $M_1$ for the reverse move.) Therefore the Jacobian factor for the Lose move from $y \rightarrow x$ is

$$\frac{|T_{y,2,v_{\text{rev}}}T_{x,1}|}{a} = \frac{\cos^2 \theta}{y_2}. \quad (3.1)$$

Plugging this into (3.1) gives a Metropolis ratio in the acceptance probability of

$$a(y, 2|x, 1) = \frac{p_{\text{lose}}(\theta; y_2)\lambda_{\text{lose}}(y_2)\cos^2 \theta}{|y_2|p_{\text{gain}}(v)\lambda_{\text{gain}}}.$$  

(5.7)
We next explain how to choose the other proposal functions to make the acceptance probability equal 1. First, we will split $p^\text{gain}(v)$ into two parts, one which steps in the normal direction, and one which steps in the tangential direction. The aim is to use the normal component $v_2 = y_2$ to balance the factor $\lambda^\text{lose}(y_2)/\lambda^\text{gain}$ as for the 1-dimensional problem in Section 5.1. These factors only depend on $y_2$, so $v_2$ can be chosen independently. The tangential component $v_1$ must also depend on $v_2$, since there is still a factor of $|y_2|$ in the denominator of (5.7) that must be balanced. Therefore we choose the density to have the form

$$p^\text{gain}(v) = p^\parallel^\text{gain}(v_1 | v_2)p^\perp^\text{gain}(v_2),$$  \hspace{1cm} (5.8)

where $p^\parallel^\text{gain}(v_2)$ is the density for the normal component of the step, and $p^\perp^\text{gain}(v_1 | v_2)$ is the density for the tangential component, given the value of the normal component.

Next, we use the choice (3.30) for $p^\text{lose}(\theta; y_2)$ and show this choice cancels the factor of $\cos^2 \theta$ in the numerator. For this example, (3.30) constructs the Lose tangent step $V = \{V_1, V_2\}$ as

$$V = \frac{-e_2 + Re_1}{-e_2 + Re_1}, \quad R \sim p_R(r),$$  \hspace{1cm} (5.9)

where $p_R(r)$ is the probability density of the random variable $R \in \mathbb{R}$, which we leave arbitrary for the moment. The inverse mapping from $V$ to $R$ takes the form

$$r(v) = \frac{v \cdot e_1}{-v \cdot e_2} = \frac{\sin \theta}{|\cos \theta|} = \frac{v_1}{v_2}. \hspace{1cm} (5.10)$$

The determinant of the jacobian of this transformation is given in (3.34) to be $|\frac{\partial r}{\partial v}| = |v - e_2|^{-d_1} = |\cos \theta|^{-2}$, so the overall density for proposing the reverse step $v^\text{rev}$ is

$$p^\text{lose}(\theta; y_2) = p^\parallel_R \left( \frac{v_{\text{rev},1}}{v_{\text{rev},2}} \right) (\cos \theta)^{-2}.$$

Substituting these specific Gain and Lose densities into (5.7) gives an acceptance probability

$$a(y, 2|x, 1) = \frac{p_R(\frac{v_1}{y_2}, \lambda^\text{lose}(y_2))}{|y_2|p^\parallel^\text{gain}(y_1 | y_2)p^\perp^\text{gain}(y_2)\lambda^\text{gain}}.$$

We have written $v$ in terms of $y_1, y_2$, and used that $v_{\text{rev},1}/v_{\text{rev},2} = y_1/y_2$.

Now, we show how to choose specific forms for these densities so the acceptance probability is 1. To balance the normal components, we can choose the densities so that

$$\lambda^\text{lose}(y_2) = p^\perp^\text{gain}(y_2)\lambda^\text{gain}.$$  \hspace{1cm} (5.12)

This has the same form as (5.3) for the 1-dimensional problem, so the same considerations apply. In particular, our choice in (5.5), where $\lambda^\text{lose}(y_2)$ is constant within an interval $y_2 \in [0, \sigma_{\text{bdy}}]$, $p^\perp^\text{gain}(y_2)$ is uniform on that same interval, and $\lambda^\text{gain} = \sigma_{\text{bdy}}\lambda^\text{lose}$, will work to satisfy the above equation.

The remaining densities must be chosen so that

$$p^\parallel^\text{gain}(y_1 | y_2) = \frac{1}{y_2} p_R \left( \frac{y_1}{y_2} \right).$$  \hspace{1cm} (5.13)

A quick calculation shows that this is possible, since $\int_{-\infty}^{\infty} p^\parallel^\text{gain}(y_1 | y_2)dy_1 = \int_{-\infty}^{\infty} p_R(r)dr = 1$, by changing variables to $r = y_1/y_2$. Therefore, once we choose a density for the tangential step of a Lose move, this sets the density for a tangential step for a Gain move via (5.13). For example, our choices from Section 3.3.2 would take the form

$$p_R(r) = \frac{1}{\sqrt{2\pi\sigma^2_{\text{tan}}}} e^{-\frac{r^2}{2\sigma^2_{\text{tan}}}}, \quad p^\parallel^\text{gain}(y_1 | y_2) = \frac{1}{\sqrt{2\pi(\sigma_{\text{tan}}y_2)^2}} e^{-\frac{y_1^2}{2(\sigma_{\text{tan}}y_2)^2}}.$$
We make a side remark that an alternative method could choose the tangential step for a Lose move in a way that depends on $y_2$, and let the tangential step for a Gain move be independent of distance. In this case the densities would have to solve
\[ p_R \left( \frac{y_1}{y_2} \bigg| y_2 \right) = y_2 p_{\text{gain}} (y_1). \]

While we haven’t tried it, we expect this to have poorer performance compared to the choice (5.13), since it implies the tangential variance for a Lose move should increase as the distance to the boundary increases, so the Lose move attempts to jump to a greater area of the boundary. For curved manifolds, this should lead to increased rejections than the method presented here, because jumps to further-away pieces of the boundary will cause the NES-L to fail more often, and will lower the acceptance probability since it will deviate even more from the flat case.

5.3 General case: $d$-dimensional manifold $\leftrightarrow$ $d + 1$-dimensional manifold

Now we show that the choice of proposal densities in Section 3.3.2 leads to 100% acceptance probability for two flat manifolds defined by affine constraints and no additional inequalities. Consider a stratification consisting of two manifolds:
\[ M_d = \{ x \in \mathbb{R}^{d+1} : q(x) = 0 \}, \quad M_{d+1} = \{ x \in \mathbb{R}^{d+1} : q(x) > 0 \}, \quad q(x) = a \cdot x + b \]  
(5.14)

where $a, b \in \mathbb{R}^{d+1}$ are constant vectors; we may assume by changing variables that $|a| = 1$. Let $I,J$ represent the labels for manifolds $M_d, M_{d+1}$ respectively. The dimensions of the manifolds are $d_I = d$, $d_J = d + 1$. We could incorporate other affine constraints that are the same for both manifolds, but by reparameterizing would obtain a case equivalent to the above. Assume the function we wish to sample is constant everywhere, $f(x) = \text{cst}$.

Consider a Gain move from $x \in M_d$ to $y \in M_{d+1}$. This is a one-sided move. The tangent step for the Gain move is $v = y - x$ and the tangent step for the reversed Lose move is $v_{\text{rev}} = (x - y)/|x - y|$. The Metropolis ratio in the acceptance probability (3.1) is calculated by substituting the densities (5.29), (3.36) (see also (3.6), (3.8), (3.18)):  
\[ a(y,J|x,I) = \frac{\lambda_{\text{lose}} f_{jl}(y) h_{jl}(x|y)}{\lambda_{\text{gain}} f_{jl}(y) h_{jl}(x|y)} = \frac{\lambda_{\text{lose}} p_{jl} \text{lose}(y_{\text{rev}})}{\lambda_{\text{gain}} p_{jl} \text{gain}(y_{\text{rev}})} \frac{|T_{y_J,v_{\text{rev}}} T_{x_I}|}{|T_{y_J,v_{\text{rev}}} T_{x_I}|^{d_I}}. \]

We know that NES, NES-L converge for any proposal when the constraints are affine.

Now calculate the geometrical factors and substitute for the tangent step densities $p_{jl} \text{gain}(y_{\text{rev}}), p_{jl} \text{lose}(y_{\text{rev}})$. We have $|T_{y_J,v_{\text{rev}}} T_{x_I}| = 1$ since the Gain move is on a flat manifold. We have $|T_{y_J,v_{\text{rev}}} T_{x_I}| = |v_{\text{rev}} \cdot a|$, the cosine of the angle between the normal vectors of subspaces $T_{y_J,v_{\text{rev}}}, T_{x_I}$. For calculating the densities, we have that $u_n = -v_{\text{opt}} = a$ since $q$ is affine (see (3.14), (3.23)). Let $v_n = v \cdot u_n$, $v_t = T_{x_I,u_n} v$ be the normal and tangential components of the Gain tangent step. The proposal is such that $v_n < \sigma_{\text{bdy}}$, so $\lambda_{\text{lose}}/\lambda_{\text{gain}} = 1/\sigma_{\text{bdy}}$. Substituting directly for all the factors above,
\[ a(y,J|x,I) = \frac{1}{\sigma_{\text{bdy}}} \frac{1}{\sqrt{2\pi\sigma_{\text{bdy}}}} e^{-\frac{|r(y_{\text{rev}})|^2}{2\sigma_{\text{bdy}}}} |v_{\text{rev}} \cdot a|^{-d_I} |v_{\text{rev}} \cdot a| |y|^{-d_I} \frac{1}{\sqrt{2\pi\sigma_{\text{bdy}}}} e^{-\frac{|y_n|^2}{2\sigma_{\text{bdy}}}} \frac{1}{\sqrt{2\pi\sigma_{\text{bdy}}}} e^{-\frac{|y_t|^2}{2\sigma_{\text{bdy}}}} \]
\[ = e^{-\frac{|r(y_{\text{rev}})|^2}{2\sigma_{\text{bdy}}}} |v_{\text{rev}} \cdot a|^{-d} |v|^{-d} \frac{v_n^d}{|y_n|^d} e^{-\frac{|y_n|^2}{2\sigma_{\text{bdy}}}}. \]

Now we use that, by geometry, $|v_n|/|v| = |v_{\text{rev}} \cdot a|$, and $|v_t|/|v_n| = |r(y_{\text{rev}})|$, to obtain that $a(y,J|x,I) = 1$.
6 Discussion and Conclusion

We have introduced a Monte Carlo method to sample systems of particles subject to constraints, such as bond distance constraints, that can break and form throughout the simulation. More generally, the method generates samples from a probability distribution defined on a stratification, a union of manifolds of different dimensions. We illustrated the method with several examples, ranging from sticky spheres, which are a model for DNA-coated colloids, to polymer physics, to calculating volumes of high-dimensional manifolds. We showed the method gives an accurate description of a system with a stiff, strong, but reversible bonds, as is the case for DNA-coated colloids.

We explored the algorithm’s efficiency, and found it to be several orders of magnitude faster than Brownian dynamics simulations for small systems characteristic of DNA-coated colloids, of around a dozen particles, but that its efficiency erodes rapidly as the number of variables $n$ of the system grows. The major cause of the slowdown is that for each proposal, the method must solve a system of nonlinear equations. If done using Newton’s method or a similar iterative solver, this requires factoring an $n \times n$ matrix several times, a calculation that costs $O(n^3)$ operations with the most generic linear algebra methods. This issue arises in any constraint-based simulation and several approaches have been developed to accelerate or parallelize solving these equations [3, 11]. It would be useful to adapt these methods to work on a stratification, or to explore sparse linear algebra techniques.

Our method is perhaps the simplest possible sampler that works for a general stratification defined by level sets of functions; it is a natural extension of a random walk Metropolis scheme on a manifold [57]. There are many ways to build upon the ideas presented here to make the method more efficient. For example, one could adapt more sophisticated sampling schemes for manifolds, such as Hamiltonian [35] or geodesic methods [32], to work on stratifications; both of these methods have velocity-like terms that can help make successive samples less correlated. In addition, one can more carefully consider how to add and subtract constraints when there are several manifolds in the stratification. Our proposal moves were carefully constructed to achieve 100% acceptance probability for two flat manifolds defined by affine constraints, but the strategy with highest acceptance probability in more general stratifications might depend on the connectivity structure of the manifolds.

An interesting extension of our method would be to generate a trajectory $X_1, X_2, \ldots$ that approximates the dynamics of a particular system. Our method is purely a sampling method, which can generate approximate samples from a probability distribution containing singular measures, but is not designed to solve a particular set of dynamical equations. However, dynamics are often important, especially for systems such as DNA-coated colloids, which diffuse rather slowly hence don’t always reach their stationary distribution on the timescale of observation. The challenge is in describing and simulating the dynamics of a diffusion process that change its intrinsic dimension, since it is not obvious how the dynamics should behave near changes in dimension. A step in this direction was taken in [6], which introduced a numerical method to simulate a “sticky Brownian motion”, a one-dimensional Brownian motion that can stick to the origin and spend finite time there. We are optimistic that the techniques from [6] can be built upon to handle higher-dimensional diffusion processes on stratifications, perhaps even by combining with tools from this paper: because our method achieves high acceptance probabilities, it already approximates some kind of dynamics.

One limitation of our method is the assumption that the constraint gradients are always linearly independent. For example, a system of $N$ sticky unit spheres can assemble into a fragment of an fcc lattice, a packing which contains nearly 6 contacts per sphere, instead of the usual 3, creating redundant constraints. While redundant constraints can in principle be removed, a more subtle issue arises when constraints become linearly dependent without introducing redundant ones; for examples where this occurs with unit sphere packings see [25]. In these cases, our algorithm will fail because we will not be able to calculate the tangent space using only gradients of the constraints, and, even if we do take a step along the tangent space, projecting back to the manifold is not always well-defined. Sometimes it is possible to deal with such degenerate configurations by formulating additional optimization problems that can be solved by semidefinite programming [24], however it is not yet clear how to build these optimization methods into our sampler.
We expect the Stratification sampler to be useful in a variety of other applications. Constraints are frequently used in biology, to model stiff bonds as well as flexible structures, but the rate-limiting step in simulations is often set by the stiffest bond that can break [43]. Sampling methods are being explored in machine learning, where constraints may offer efficient ways to explore parameter spaces of interest [33]. Our method may also generate interesting data for problems in discrete geometry; one example is to ask which kinds of graphs may be formed from packing spheres together [9]. We hope the method can be adapted to these kinds of systems, and others yet to be imagined.

Acknowledgements

Thank you to Jonathan Goodman, Jeff Cheeger, and Mark Goresky for helpful discussions regarding sampling and stratifications, to Nawaf Bou-Rabee for carefully reading early drafts of the paper, and to Anthony Trubiano for pointing out bugs in an earlier version of the code. I would also like to acknowledge support from the US Department of Energy [de-sc0012296] and the Alfred P. Sloan Foundation.

A Appendix: Verifying the invariant measure

In this section we show that the measure \( \rho \) defined in (2.11) is an invariant measure for the Markov chain \( X_1, X_2, \ldots \) constructed in Section 3 if \( X_k \sim \rho \), then \( X_{k+1} \sim \rho \). This argument is very similar to that in Green [20] Section 3.2, which shows that RJMC satisfies detailed balance, the difference being that Green [20] considers a stratification formed from a union of Euclidean spaces, whereas we consider a union of embedded manifolds. We include the argument nonetheless for completeness.

To start, define the product space \( \mathcal{C} = \mathcal{S} \times \mathcal{J} \). It is convenient to change notation slightly and let \( \bar{\rho} \) be the measure in (2.11), and define \( \rho(dx, I) = Z^{-1} f_I(x) \mu_I(dx) \) to be the desired joint probability distribution to find the system at configuration \( x \in \mathcal{S} \) and with labels \( I \in \mathcal{J} \). The measure in (2.11) is obtained by summing over labels as \( \bar{\rho}(dx) = \sum_{I \in \mathcal{I}} \rho(dx, I) \). We will show that (using our new notation)

\[
(X_k, I_k) \sim \rho \quad \Rightarrow \quad (X_{k+1}, I_{k+1}) \sim \rho, \tag{A.1}
\]

which implies the parallel result for \( \bar{\rho} \).

Let \( x = X_k, I = I_k \). The probability to successfully propose \((y, J)\) starting at \((x, I)\) has distribution

\[
P(dy, J|x, I) = \lambda_{ij}(x) h_{ij}(y|x) a(y, J|x, I) \mu_J(dy) + \xi_{ij}(x) \delta_i(y - x) \mu_J(dy). \tag{A.2}
\]

The first term in the sum is the probability distribution for proposing \((y, J)\) and then accepting it; the second term is the probability distribution for remaining at \((x, I)\). The latter is a product of \( \xi_{ij}(x) \), the probability of remaining at the starting point \((x, I)\) (either because of failing to produce a proposal point, or because the proposal point was rejected), and a measure \( \delta_i(y - x) \mu_J(dy) \). We write \( \delta_i(y - x) \) to mean the delta-function with respect to the measure \( \mu_i \); it is defined so that, for any integrable function \( g : M_i \to \mathbb{R} \), we have \( \int_{y \in M_i} g(y) \delta_i(y - x) \mu_j(dy) = g(x) \) if \( I = J \), and \( \int_{y \in M_i} g(y) \delta_i(y - x) \mu_j(dy) = 0 \) otherwise.

Now suppose that \((X_k, I_k) \sim \rho \). Let \( \rho' \) be the distribution of \((X_{k+1}, I_{k+1})\). It is computed as

\[
\rho'(dy, J) = Z^{-1} \sum_{I \in \mathcal{J}} \int_{x \in \mathcal{S}} P(dy, J|x, I) \rho_I(dx)
\]

\[
= Z^{-1} \sum_{I \in \mathcal{J}} \int_{x \in M_I} \left( \lambda_{ij}(x) h_{ij}(y|x) a(y, J|x, I) \mu_J(dy) + \xi_{ij}(x) \delta_i(y - x) \mu_j(dy) \right) f_I(x) \mu_I(dx)
\]

\[
= Z^{-1} \mu_J(dy) \left( \sum_{I \in \mathcal{J}} f_I(x) \lambda_{ij}(x) h_{ij}(y|x) a(y, J|x, I) \mu_I(dx) + \xi_{ij}(y) f_J(y) \right). \tag{A.3}
\]
In the last step we used \( \int_{x \in M_i} f_j(x) \xi_i(x) \delta_j(y-x) \mu_j(dx) = f_j(y) \xi_j(y) \), and then we used that \( \sum_{i=1} f_j(y) \xi_i(y) \mu_j(dy) = f_j(y) \xi_j(y) \mu_j(dy) \), since measure \( \mu_j(y) \) only gives nonzero weight to \( y \in M_j \), and if \( y \in M_j \) and \( I \neq J \) then \( \xi_i(y) = 0 \), by definition.

Now, suppose that the acceptance probability \( a \) satisfies the following relation:

\[
f(x) \lambda_{JJ}(x) h_{JJ}(y|x)a(y,J|x,I) = f(y) \lambda_{JJ}(y) h_{JJ}(x|y)a(x,I|y,J).
\]

It is straightforward to verify that the particular choice of \( a \) in (3.1) satisfies this. Then, substituting for \( a(y,J|x,I) \) in (A.3) gives

\[
\rho'(dy,J) = Z^{-1} f_j(y) \mu_j(dy) \left( \sum_{I \in \mathcal{J}} \int_{y \in M_i} \lambda_{JJ}(y) h_{JJ}(x|y)a(x,I|y,J) \mu_i(dx) + \xi_j(y) \right).
\]

As a final step we rewrite function \( \xi_j(y) \) in terms of the other ingredients of the proposal distribution. This function, the total probability of remaining at the starting point \((y,J)\), equals the probability of failing to produce a proposal, plus the probability of producing a proposal but rejecting it. Since the only other possibility is to to produce a proposal and accept it, we must have that \( \xi_j(y) \) plus the probability of producing a successful proposal equals 1:

\[
\xi_j(y) = 1 - \sum_{I \in \mathcal{J}} \int_{x \in M_i} \lambda_{JJ}(y) h_{JJ}(x|y)a(x,I|y,J) \mu_i(dx).
\]

Substituting (A.6) into (A.5) gives

\[
\rho'(dy,J) = Z^{-1} f_j(y) \mu_j(dy),
\]

which is the desired result.

**B Appendix: Summary of the algorithm**

This section provides pseudocode, written at a high level to show the main steps of the algorithm. The pseudocode depends on having first constructed a stratification, a topic we comment on in Section B.1 before describing the main algorithms provided in the pseudocode in Section B.2. Code used to run our examples is available at [1].

The algorithm depends on 5 parameters: \( \sigma, \lambda_{\text{lose}}, \lambda_{\text{gain}}, \sigma_{\text{bdy}}, \sigma_{\text{tan}} \) (4 parameters if \( \lambda_{\text{gain}} \) is set via (3.18), but we keep it as an independent parameter.) These parameters could depend on the manifold, but in our implementation we choose them to be independent of manifold. If one wishes to make the parameters manifold-dependent, then \( \sigma \) may be different for each manifold, and \( \lambda_{\text{lose}}, \lambda_{\text{gain}}, \sigma_{\text{tan}}, \sigma_{\text{bdy}} \) may be different for each pair of manifolds.

**B.1 Constructing a Stratification**

To form a stratification we need a set of functions \( \mathcal{Q} \) and a set of labels \( \mathcal{J} \). Here is how we represent these on a computer.

We start from a list of functions \( \mathcal{Q} = \{ q_1, \ldots, q_{n_{\text{fcns}}} \} \) and their gradients \( \{ \nabla q_1, \ldots, \nabla q_{n_{\text{fcns}}} \} \), where \( q_i : \mathbb{R}^n \rightarrow \mathbb{R} \) and \( n_{\text{fcns}} = |\mathcal{Q}| \). These are implemented by providing functions \( \text{eq}(i,x) \) and \( \text{jac}(i,x) \) defined so that \( q_i(x) = \text{eq}(i,x), \nabla q_i(x) = \text{jac}(i,x) \). Both \( \text{eq}, \text{jac} \) are application-dependent; to change the application one simply needs to rewrite these functions (and change the number of variables \( n \) and number of functions \( n_{\text{fcns}} \)).

The labels for a given manifold in the stratification are represented with vector

\[
L \in \{ C_{\text{eq}}, C_{\text{in}}, C_{\text{tan}} \}^{n_{\text{fcns}}}
\]
where $C_{eq} = 1$, $C_{in} = 2$, $C_{none} = 0$ are constants labelling whether each function is a constraint, inequality, or neither, respectively. For example, the labels $L = (C_{eq}, C_{eq}, C_{none}, C_{in})$ represent the manifold $M_L = \{x \in \mathbb{R}^n : q_1(x) = 0, q_2(x) = 0, q_4(x) > 0\}$. We change notation slightly in this section, and use the vector $L$ instead of the corresponding subset $I$ to represent labels for manifolds and tangent spaces.

Recall that for a given manifold $M_L$, the number of equations that define it is written as $m_L$, and its intrinsic dimension is $d_L$. The number of equations is computed as $m_L = \#i : L_i = C_{eq}$ and $d_L = n - m_L$ by our assumption that the gradients of the constraints are linearly independent everywhere on $M_L$.

To finish constructing a stratification we need a set of labels $I$ indicating which manifolds are in the stratification. We do this in one of two ways: either by providing a list of labels in advance, or by forming $I$ from all subsets of labels obtained by letting certain functions vary between constraint/inequality. We distinguish these cases in the code by the parameter $nmanif$. When $nmanif > 0$ we use the first method, and $nmanif = |\mathcal{I}|$ represents the total number of manifolds in the stratification. When $nmanif = 0$, we use the second method, which doesn’t require forming a complete list of manifolds.

The overall goal of defining $I$ is to access $N_{gain}(L), N_{lose}(L)$, the set of Gain and Lose neighbours for a manifold $M_L$ defined by labels $L$, when requested. How we compute the neighbours depends on which method is used to construct $I$.

In the first case, when $nmanif > 0$, we provide a list of labels in the form of a matrix, $\text{Llist}$, whose $i$th row $L(i) = \text{Llist}(i,:)$ gives the labels for the $i$th manifold in the stratification. The algorithm pre-computes $N_{gain}(L(i)), N_{lose}(L(i))$ by checking condition (2.13). That is, given $L(i) = \text{Llist}(i,:)$, the algorithm checks whether for each label $L(i) = \text{Llist}(j,:), j \neq i, L(i)$ differs from $L(i)$ by at most one equation. The sets $N_{gain}(L(i)), N_{lose}(L(i))$ are stored as lists of indices.

In the second case, when $nmanif = 0$, the algorithm computes $N_{gain}(L), N_{lose}(L)$ on the fly. In the setup we must provide a set of flags $\text{FixFcns} \in \{\text{fix}, \text{vary}\}^{n\text{manif}}$ with $\text{fix} = 1, \text{vary} = 0$, such that $\text{FixFcns}(i) = \text{fix}$ if the label for function $q_i$ cannot change from what is provided initially, and $\text{FixFcns}(i) = \text{vary}$ if the label for function $q_i$ can vary between constraint/inequality. Then, given labels $L$, the algorithm constructs $N_{gain}(L)$ by considering all possible ways to turn a single inequality into an equation (by performing a single flip $C_{eq} \rightarrow C_{in}$ for each function that is allowed to vary), and it constructs $N_{lose}(L)$ by considering all possible ways to turn a single equation into an inequality (by performing a single flip $C_{in} \rightarrow C_{eq}$ for each function that is allowed to vary.)

### B.2 Pseudocode

The main algorithm is \texttt{SampleStrat} (Algorithm 1). This takes as input a state $(X_n, L_n)$ representing the configuration $X_n \in \mathbb{R}^n$ and labels $L_n \in \{C_{eq}, C_{in}, C_{none}\}^{n\text{manif}}$ of the $n$th step of the Markov chain, and outputs the $n+1$th state $X_{n+1}, L_{n+1}$. This algorithm in turn calls \texttt{LamPropose} (Algorithm 2) to generate a proposal label $L$ and \texttt{VPropose} (Algorithm 3) to generate a proposal step $\nu$ in the tangent space. It then calls \texttt{TakeStep} (Algorithm 4) to move in the direction $\nu$ and project back to the manifold, using projection methods NES or NES-L (also Algorithm 4) to obtain a proposal $y \in M_L$. \texttt{SampleStrat} then performs a variety of checks on the proposal $y, L$ to determine whether to accept it or reject it.

Most of these algorithms require information about the tangent space to a manifold, in the form of a matrix $T_{x, L}$, or sometimes $T_{x, L, v}$. Recall that $T_{x, L}, T_{x, L, v}$ are matrices whose columns form an orthonormal basis of spaces $T_{x, L}, T_{x, L, v}$ respectively, spaces which are defined in Section 3.2. Algorithm 5 shows how these matrices are calculated, using the QR decomposition of a matrix of gradients of the form (2.8). Once the matrices have been calculated for a state $x, L$, they are stored together with that state and reused wherever needed.

We remark that NES and NES-L perform essentially the same calculation, namely they solve a system of equations $q_k(x + Qa) = 0$ for $k \in I$ where $I$ is some subset of $\{1, \ldots, n\text{fcns}\}$. The only differences between the algorithms are (i) they use a different initial condition for Newton’s method, and (ii) the ordering of the constraints is different. We prefer to keep these functions separated to make it easier to interpret their output.

In the pseudocode that follows, we don’t explicitly write out all the variables that must be passed to each
function. Our implementation makes heavy use of objects, so we can efficiently pass all the information associated with a state $x, L$, such as $T_{x,L}$, $N_{\text{gain}}(L)$, $N_{\text{lose}}(L)$, $m_L$, $d_L$, and the values of the equations, inequalities, and gradients of the equations. We use the function argument $(x, L)$ somewhat loosely to mean all information associated with that particular state that has been calculated so far.
Algorithm 1

Given \( x = X_n, L_n \), generate the next point \( X_{n+1}, L_{n+1} \) in the Markov chain.

1. Parameters: \( \sigma, \lambda_{\text{lose}}, \lambda_{\text{gain}}, \sigma_{\text{bdy}}, \sigma_{\text{tan}} \)
2. procedure \text{SAMPLESTRAT}(x = X_n, L_n)
   3. \textbf{Proposal:}
      \( \triangleright \) Generate a proposal \( y, L \) and movetyperev \( \in \{\text{Same}, \text{Gain}, \text{Lose}\} \)
      movetyperev, \( L = \text{LAMPROPOSE}(x, L_0) \)  \( \triangleright \) Propose type of move and new labels
      5. \( v = \text{VPROPOSE}(x, L_0, \text{movetyperev}, L) \) \( \triangleright \) Propose step in tangent space
      6. \( y, \text{newtonflag}, \alpha = \text{TAKESTEP}(x, L_n, \text{movetyperev}, L, v) \)
         \( \triangleright \) Takes a step \( v \) in the tangent space starting at \( x \) and projects to manifold \( M_n \).
         If movetyperev \( \in \{\text{Same, Gain}\} \), solves for \( y \in M_n \) s.t. \( y = x + v + w, w \perp T_{x,L} \)
         If movetyperev \( = \text{Lose} \), solves for \( \alpha \in \mathbb{R}, y \in M_n \) s.t. \( y = x + \alpha v + w, w \perp T_{x,L} \)
   8. \textbf{Projection Check:}
      9. if newtonflag \( = \text{fail} \) then \( \triangleright \) Projection step (NES or NES-L) failed to converge
         Reject proposal: \( X_{n+1} = X_n, L_{n+1} = L_n \). Return.
      10. if movetyperev \( = \text{Lose and } \alpha < 0 \) then \( \triangleright \) Lose move went in opposite \( v \) direction
          Reject proposal: \( X_{n+1} = X_n, L_{n+1} = L_n \). Return.
   13. \textbf{Inequality Check:}
      14. if \( q_i(y) < 0 \) for some \( i \) s.t. \( L(i) = C_{\text{in}} \) then
          Reject proposal: \( X_{n+1} = X_n, L_{n+1} = L_n \). Return.
   16. \textbf{Metropolis-Hastings Step:}
      17. Compute \( \lambda_{\text{same}}(x), \lambda_{\text{gain}}(x), \lambda_{\text{lose}}(x); \lambda_{\text{same}}(y), \lambda_{\text{gain}}(y), \lambda_{\text{lose}}(y) \) via \( \text{(3.19)}, \text{(3.17)} \)
      18. Get densities \( \lambda_{ij}(x) h_{ij}(y|x) \) for forward move via \( \text{(3.22)}, \text{(3.29)}, \) or \( \text{(3.36)} \)
      19. Set movetyperev \( = \text{Same, Lose, Gain} \) for movetyperev \( = \text{Same, Gain, Lose} \) respectively
      20. if movetyperev \( = \text{Same or Gain} \) then \( \triangleright \) Construct steps for reverse move
         Find \( v' \in T_{x,L_n} \) such that \( x = y + v' + w' \perp T_{y,L_n} \) \( \triangleright \) See \( \text{(3.5)} \)
      22. else if movetyperev \( = \text{Lose} \) then
         Find \( v' \in T_{y,L_n} \) such that \( x = y + v' + w' \perp T_{y,L_n} \) \( \triangleright \) Similar to \( \text{(3.5)} \)
         \( \alpha' = |v'|, v' \leftarrow v'/|v'| \)
      25. Get densities \( \lambda_{ij}(y) h_{ij}(x|y) \) for reverse move via \( \text{(3.22)}, \text{(3.29)}, \) or \( \text{(3.36)} \), using \( v', \alpha' \)
         \( \triangleright \) Temporarily set accessibility factor \( 1_{\text{movetyperev}(y)}(x) = 1 \)
      27. Compute acceptance probability \( \alpha = a(y, L|x, L_n) \) via \( \text{(3.1)} \) \( \triangleright \) Also requires \( f(x), f(y) \)
      28. if \( U \sim \text{Unif([0,1])} > \alpha \) then
         Reject proposal: \( X_{n+1} = X_n, L_{n+1} = L_n \). Return.
   30. \textbf{Reverse Projection:}
      31. \( x', \text{newtonflagrev}, \alpha' = \text{TAKESTEP}(y, L, \text{movetyperev}, L, \nu') \)
         \( \triangleright \) Takes a step \( v' \) in the tangent space starting at \( y \) and projects to manifold \( M_n \).
         If movetyperev \( \in \{\text{Same, Gain}\} \), solves for \( x' \in M_n \) s.t. \( x' = y + v' + w', w' \perp T_{y,L_n} \)
         If movetyperev \( = \text{Lose} \), solves for \( \alpha', x' \in M_n \) s.t. \( x' = y + \alpha' v' + w', w' \perp T_{y,L_n} \)
      33. if newtonflag \( = \text{fail} \) then \( \triangleright \) Reverse projection step failed to converge
         Reject proposal: \( X_{n+1} = X_n, L_{n+1} = L_n \). Return.
      35. if \( x' \neq x \) then \( \triangleright \) Projection converged, but to wrong point
         Reject proposal: \( X_{n+1} = X_n, L_{n+1} = L_n \). Return.
      37. if movetyperev \( = \text{Lose and } \alpha' < 0 \) then
         Reject proposal: \( X_{n+1} = X_n, L_{n+1} = L_n \). Return.
      39. Accept proposal: \( X_{n+1} = y, L_{n+1} = L \). Copy \( T_{y,L}, N_{\text{gain}}(L), N_{\text{lose}}(L) \) to \( X_{n+1} \). Return.
40. \textbf{end procedure}
Algorithm 2 Propose a type of move and new labels

1: Parameters: \( \lambda_{\text{lose}}, \lambda_{\text{gain}}, \sigma_{\text{bdy}} \)
2: procedure \textsc{LAMPropose}(x, L_0) 
3: \begin{align*}
&\text{Get } N_{\text{gain}}(L_0), N_{\text{lose}}(L_0) \quad \triangleright \text{Lists of labels, e.g. } N_{\text{lose}}(L_0) = \{L'_1, L'_2, \ldots, L'_{\vert N_{\text{lose}}(L_0)\vert}\} \\
&\text{\quad \triangleright \text{If } n_{\text{manif}}>0 \text{ these lists are pre-computed for each manifold; if } n_{\text{manif}}=0 \text{ they are obtained by adding each inequality or subtracting each constraint to current labels}} \\
&\text{Compute nearby Lose neighbours as in (3.16):} \\
&\quad N_{\sigma_{\text{bdy}} \text{lose}}(L_0, x) \\
&\quad \text{\ for } i \leftarrow 0, i < \vert N_{\text{lose}}(L_0)\vert \text{ do} \\
&\quad \quad \text{if } \text{DISTMIN}(x, L_0, k_i^{\text{(change)})} \text{ then} \\
&\quad \quad \quad N_{\text{lose}}(L_0, x) \leftarrow N_{\sigma_{\text{bdy}} \text{lose}}(L_0, x) \cup \{L'_i\} \\
&\quad \end{align*}

5: \begin{align*}
&\quad \text{end for} \\
&\quad n_{\text{gain}} = \vert N_{\text{gain}}(L_0)\vert, n_{\text{lose}} = \vert N_{\sigma_{\text{bdy}} \text{lose}}(L_0, x)\vert \\
&\quad \triangleright \text{See (3.17)} \\
&\text{Compute } \Lambda_{\text{same}}(x), \Lambda_{\text{gain}}(x), \Lambda_{\text{lose}}(x) \text{ via (3.19):} \\
&\quad \Lambda_{\text{gain}}(x) = \lambda_{\text{gain}}, \Lambda_{\text{lose}}(x) = \lambda_{\text{lose}} \\
&\text{\ if } n_{\text{gain}} == 0 \text{ then } \Lambda_{\text{gain}}(x) = 0 \\
&\text{\ if } n_{\text{lose}} == 0 \text{ then } \Lambda_{\text{lose}}(x) = 0 \\
&\quad \Lambda_{\text{same}}(x) = 1 - \Lambda_{\text{gain}}(x) - \Lambda_{\text{lose}}(x) \\
&\text{Choose a move with the desired probabilities:} \\
&\text{\ if } U < \Lambda_{\text{same}}(x) \text{ then} \\
&\quad \text{movetype } = \text{Same} \\
&\quad L_1 = L \\
&\text{\ else if } U < \Lambda_{\text{same}}(x) + \Lambda_{\text{gain}}(x) \text{ then} \\
&\quad \text{movetype } = \text{Gain} \\
&\quad L_1 \sim \text{Uniform}(N_{\sigma_{\text{bdy}} \text{lose}}(L_0, x)) \\
&\text{\ else} \\
&\quad \text{movetype } = \text{Lose} \\
&\quad L_1 \sim \text{Uniform}(N_{\text{lose}}(L_0, x)) \\
&\quad \triangleright \text{Choose labels uniformly from nearby Lose neighbours} \\
&\text{return movetype, } L_1 \\
&\text{end procedure} \\

30: \text{Estimate the minimum distance from } x \text{ to boundary } \{z : q_i(z) = 0\} \text{ over all directions } v \in T_{x,i}. \\
31: \text{function \textsc{DISTMIN}(x, L, k)} \\
32: \quad \text{return } |q_i(x)|/|T_{x,i} \nabla q_i(x)| \\
33: \text{\triangleright \text{Estimated by linearizing } q_i, \text{ as in (3.15)}} 
34: \text{end function}
Algorithm 3 Propose a step $v$ in the tangent space

1: Parameters: $\sigma$, $\sigma_{\text{bdy}}, \sigma_{\text{tan}}$

2: procedure \textsc{VPROPOSE}(x, $L_0$, movetype, $L_1$)

3: \textbf{if} movetype == Same \textbf{then}

4: \hspace{1em} if $d_{L_0} == 0$ then $v = (0, \ldots, 0) \in \mathbb{R}^n$ \hspace{1em} $\triangleright$ Dimension of $M_{L_0}$ is 0; remain on current point

5: \hspace{1em} else

6: \hspace{2em} Generate $r_i \sim \sigma \cdot N(0, 1)$ for $i = 1, \ldots, d_{L_0}$ \hspace{1em} $\triangleright$ $r$ is vector of i.i.d. normals

7: \hspace{2em} $v = T_{x, L_0} r$ \hspace{1em} $\triangleright$ Step in tangent space $T_{x, L_0}$

8: \textbf{end if}

9: \textbf{if} movetype == Gain \textbf{then}

10: \hspace{1em} Let $i_{\text{change}}$ be index of constraint removed from $L_0$ to $L_1$

11: \hspace{2em} Calculate $T_{x, L_1}$ \hspace{1em} $\triangleright$ Basis for tangent space with $q_{\text{change}}$ removed

12: \hspace{2em} $u_n = T_{x, L_0} T_{x, L_1}^T \nabla q_{\text{change}}, \quad u_n \leftarrow u_n / |u_n|$ \hspace{1em} $\triangleright$ See (3.23)

13: \hspace{2em} Generate $U \sim \text{Unif}([0, 1])$

14: \hspace{2em} if move is one-sided then $v_n = \sigma_{\text{bdy}} U$ \hspace{1em} $\triangleright$ Component of step in normal direction

15: \hspace{2em} if move is two-sided then $v_n = \sigma_{\text{bdy}} (2U - 1)$

16: \hspace{2em} $v = v_n u_n$ \hspace{1em} $\triangleright$ Step in normal direction

17: \hspace{2em} if $d_{L_0} \geq 1$ then \hspace{1em} $\triangleright$ There exist tangential directions

18: \hspace{2em} Generate $r_i \sim \sigma_{\text{tan}} u_n \cdot N(0, 1)$ for $i = 1, \ldots, d_{L_0}$ \hspace{1em} $\triangleright$ Components of tangential steps

19: \hspace{2em} $v \leftarrow v + T_{x, L_0} u_n r$ \hspace{1em} $\triangleright$ Add tangential steps to $v$; see (3.30)

20: \textbf{end if}

21: \textbf{if} movetype == Lose \textbf{then}

22: \hspace{1em} Let $i_{\text{change}}$ be index of constraint added from $L_0$ to $L_1$

23: \hspace{2em} $u_n = T_{x, L_0} T_{x, L_1}^T \nabla q_{\text{change}}, \quad u_n \leftarrow u_n / |u_n|$ \hspace{1em} $\triangleright$ $u_n = v_{\text{opt}}$, as in (3.14)

24: \hspace{2em} if move is two-sided and $q_{\text{change}}(x) < 0$ then $u_n \leftarrow -u_n$

25: \hspace{2em} $v = -u_n$ \hspace{1em} $\triangleright$ Normal component of step

26: \hspace{2em} if $d_{L_0} \geq 2$ then \hspace{1em} $\triangleright$ Tangential components of step

27: \hspace{2em} Construct $T_{x, L_0} u_n$

28: \hspace{2em} Generate $r_i \sim \sigma_{\text{tan}} \cdot N(0, 1)$ for $i = 1, \ldots, d_{L_0} - 1$ \hspace{1em} $\triangleright$ See (3.30)

29: \hspace{2em} $v \leftarrow v + T_{x, L_0} u_n r$

30: \hspace{2em} $v \leftarrow v / |v|$ \hspace{1em} $\triangleright$ Send back direction only

31: \textbf{end if}

32: return $v$

33: \textbf{end procedure}
Algorithm 4 Take a step \( v \) in the tangent space from \( x \) and project back to manifold

1: Parameters: \( \text{tol}, \text{MaxIter} \)
2: procedure \textsc{TakeStep}(\( x, L_0, \text{movetype}, L_1, v \))
3: \hspace{1em} if \( \text{movetype} \in \{\text{Same, Gain}\} \) then
4: \hspace{2em} \( z = x + v \) \hfill \( \triangleright m_{L_1} = \text{number of equations in } L_1 \)
5: \hspace{2em} if \( m_{L_1} > 0 \) then \hfill \( \triangleright \) Compute \( Q_{L_1} \) via (2.8)
6: \hspace{3em} \( y, \text{newtonflag} = \text{NES}(z, Q_{L_1}, L_1) \) \hfill \( \triangleright \) Project back to manifold
7: \hspace{2em} else \hfill \( \triangleright \) No projection needed if \( M_{L_1} \) is defined only by inequalities
8: \hspace{3em} \( y = z, \text{newtonflag} = \text{success} \)
9: \hspace{1em} else if \( \text{movetype} = \text{Lose} \) then
10: \hspace{2em} Construct \( Q^{(i)} = (Q_{L_0} | v) \), with \( Q_{L_0} \) as in (2.8)
11: \hspace{3em} Let \( i_{\text{change}} \) be index of constraint added from \( L_0 \) to \( L_1 \) \hfill \( \triangleright q_{\text{change}} \) is added equation
12: \hspace{3em} \( y, a, \text{newtonflag} = \text{NES-L}(x, Q^{(i)}, L_0, i_{\text{change}}, v) \) \hfill \( \triangleright \) Modified projection for a Lose move
13: \hspace{1em} return \( y, \text{newtonflag}, (a) \)
14: end procedure
15: function \text{NES}(z, Q, L)
16: \hspace{1em} Set \( a = (0, 0, \ldots, 0) \in \mathbb{R}^n_l \) \hfill \( \triangleright \) Deterministic initial condition
17: \hspace{2em} for \( i = 0, i < \text{MaxIter} \) do
18: \hspace{3em} \( F \leftarrow (q_k(z + Qa))_{k:L(k) = C_{eq}} \) \hfill \( \triangleright \) Vector containing current values of equations
19: \hspace{3em} if max, \( |F_j| < \text{tol} \) then \hfill \( \triangleright \) Success! Solution converged
20: \hspace{4em} \( y = z + Qa \)
21: \hspace{3em} return \( y, \text{newtonflag} = \text{success} \)
22: \hspace{3em} \( \hat{Q} = (\nabla q_k(z + Qa))_{k:L(k) = C_{eq}} \) \hfill \( \triangleright \) Current value of constraint gradients in \( L_1 \) as in (2.8)
23: \hspace{3em} \( J = \hat{Q}^T Q \) \hfill \( \triangleright \) Jacobian of system to be solved
24: \hspace{3em} Solve \( J \Delta a = -F \) for \( \Delta a \) \hfill \( \triangleright \) We used LU decomposition with partial pivots
25: \hspace{3em} \( a \leftarrow a + \Delta a \)
26: end for
27: return \( y = \text{NaN}, \text{newtonflag} = \text{fail} \) \hfill \( \triangleright \) If we got this far, solution didn't converge
28: end function

19: function \text{NES-L}(x, Q, L_0, L_1, v)
30: \hspace{1em} Set \( a = (0, 0, \ldots, 0, h) \in \mathbb{R}^n_0 \) \hfill \( \triangleright \) Linearizing about 0 gives \( q_i(hv) = 0 + O(h^2) \)
31: \hspace{2em} for \( i = 0, i < \text{MaxIter} \) do
32: \hspace{3em} \( F \leftarrow (q_k(x + Qa))_{k:L_0(k) = C_{eq}, k = i_0} \) \hfill \( \triangleright \) Current values of equations in \( L_0 \) and \( i_0 \)
33: \hspace{3em} if max, \( |F_j| < \text{tol} \) then \hfill \( \triangleright \) Success! Solution converged
34: \hspace{4em} \( y = x + Qa \)
35: \hspace{4em} return \( y, \text{newtonflag} = \text{success} \) \hfill \( \triangleright \) \( a \) is the last element of \( a \)
36: \hspace{3em} \( \hat{Q} = (\nabla q_k(x + Qa))_{k:L_0(k) = C_{eq}, k \neq i_0} \) \hfill \( \triangleright \) (2.8) for \( L_0 \) with additional column
37: \hspace{3em} \( J = \hat{Q}^T \hat{Q} \) \hfill \( \triangleright \) Jacobian of system to be solved
38: \hspace{3em} Solve \( J \Delta a = -F \) for \( \Delta a \) \hfill \( \triangleright \) We used LU decomposition with partial pivots
39: \hspace{3em} \( a \leftarrow a + \Delta a \)
40: end for
41: return \( y = \text{NaN}, \text{newtonflag} = \text{fail} \) \hfill \( \triangleright \) If we got this far, solution didn't converge
42: end function
Algorithm 5 Calculate an orthonormal basis of a tangent space

1. Calculates $T = T_{x,L}$, a matrix whose columns form an orthonormal basis of $\mathcal{T}_{x,L}$.
2. function $\text{TAN}(x, L)$
3.   if $0 < d_L < n$ then
4.     Calculate $Q_L$ in (2.8)
5.     $W = QR(Q_L)$ → QR decomposition
6.     $W$ has block form $W = (N | T)$, where $N \in \mathbb{R}^{n \times m_L}$ is an orthonormal basis of $N_{x,L}$, and $T = T_{x,L} \in \mathbb{R}^{n \times d_L}$ is an orthonormal basis of $\mathcal{T}_{x,L}$
7.     Set $T$ to be the last $d_L$ columns of $W$
8.   else if $d_L == n$ then → No equations; moving in ambient Euclidean space
9.     $T = I \in \mathbb{R}^{n \times n}$ → The $n \times n$ identity matrix
10.   else if $d_L == 0$ then → We're on a 0-dimensional point
11.     Don't need $T$, since there is no tangent space.
12.   end if
13. return $T$
14. end function

15. Calculates $T_v = T_{x,L,v}$, a matrix whose columns form an orthonormal basis of $\mathcal{T}_{x,L,v}$.
16. The function assumes that $v \in \mathcal{T}_{x,L}$; if not, the function should be modified.
17. function $\text{TANV}(x, L, v)$
18.   if $d_L \geq 2$ then
19.     $T = \text{TAN}(x, L)$
20.     $P = I - vv^T / |v|^2$, here $I \in \mathbb{R}^{n \times n}$ is identity matrix
21.     $P$ is the orthogonal projection matrix onto the space perpendicular to $v$
22.     $W = QR(P \cdot T)$ → QR decomposition
23.     Set $T_v$ to be the first $d_L - 1$ columns of $W$.
24.   else → We're on a point or a line
25.     Don't need $T_{x,L,v}$ since there's at most 1 direction
26.     return $T_v$
27. end function
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