

1. Designing a simple model system to test the validity of individual MC moves

All other energy scales are set to zero except for the anisotropic bond energy scale. The simulation box size is set to $3 \times 3 \times 3$ to ensure that all beads are neighbors under periodic boundary conditions. Therefore, it is crucial that all positional configurations permit all possible configurations of rotational bond formation. We can determine the multiplicity of a state with a specific energy value by considering the possible combinations of exclusive bonds (Table 2). Furthermore, all steps could be assumed to be in equilibrium because of the small size of the system. The parameters used in this paper are listed in Table 1.

MC_TEMP	1.0
N_STEPS	$1.0 * 10^6$
BOX_SIZE	$3 * 3 * 3$

Table 1. parameters for the model system

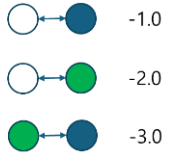


E scales	System type	Results					
		E	0	-1.0	-2.0	-3.0	-4.0
		$m(E)$	1	6	9	14	12
		E	0	-1.0	-2.0	-3.0	
		$m(E)$	1	9	18	6	

Table 2. $m(E)$ denotes a multiplicity for a positional configuration which total energy is E . $m(E)$ can be calculated by counting all possible combinations for rotational bond configurations.

2. An Error in calculating Rosenbluth weight factor for a single bead

As introduced in the original paper, each MC move in LaSSI samples the resulting state according to the Boltzmann factor of the rotational energy. While conducting each MC move, a next state suggested and accepted according to their characteristic rule designed to ensure the microscopic reversibility. The Rosenbluth weight factor of a monomer is defined as the sum of the Boltzmann factors of all possible rotational states of the monomer, and it is used to calculate a biased sampling ratio and corresponding acceptance ratio. However, discrepancies were found between the main text's explanation and the code implementation, leading to violations of detailed balance, which were not intended.

2.1. Original workflow

A monomer can be in an associated or a dissociated state, with a specified binding partner in the

associated state. This defines the rotational state of a monomer. To change the rotational state, all 26 adjacent sites to a monomer are exhaustively sampled to construct a list of potential binding partners that are not bonded with another bead. The number of potential binding partners is denoted by b . The rotational state of the monomer is changed at random by the following steps:

1. Draw a random number k from a uniform distribution between $[0,1]$.
2. If $k \leq 1/(b+1)$, the monomer is set to be in a dissociated state.
3. If $k > 1/(b+1)$, the j -th candidate bond is formed by drawing a random number j from the distribution $P_j = e^{-\beta\epsilon_j}/W$, where ϵ_j denotes the energy of the j -th candidate bond and W denotes the Rosenbluth weight factor of the monomer, which defined as $W = \sum_{j=1}^{j=b} e^{-\beta\epsilon_j}$.

The ratio between an associated state and a dissociated state did not agree with the ratio of the corresponding Boltzmann factors on this procedure. Furthermore, this inconsistency was not properly handled in calculating the acceptance ratio of the corresponding MC move. Additionally, there was an error in the acceptance ratio calculation, as the total energy of each state was used instead of just the positional energy from correct equation (Eq. 1). These errors resulted in an inappropriate equilibrium distribution except for the special case where all monomers have identical energy.

2.2. Corrected workflow

A list of potential binding partners is constructed using the same procedure. However, a dissociated state is considered as a 0th rotational state with an energy value of 0. Thus, a calling for random number is conducted once, and the Rosenbluth weight factor is defined as $W = \sum_{j=0}^{j=b} e^{-\beta\epsilon_j}$, not a $\sum_{j=1}^{j=b} e^{-\beta\epsilon_j}$. Consequently, we can use the acceptance ratio determined by Equation (1) as suggested in the previous paper, with corrected W_i and W_j .

$$A_{ij} = \min\left\{1, \frac{W_j}{W_i} \exp[-\beta(E_{j,pos} - E_{i,pos})]\right\} \quad (1)$$

2.3. Impact on the result energy distribution

Detailed balance was broken in the original version of LaSSI, causing discrepancies in the energy distributions compared to the ideal distribution. Corresponding ideal energy distributions for a model system were calculated as described in Section 1.

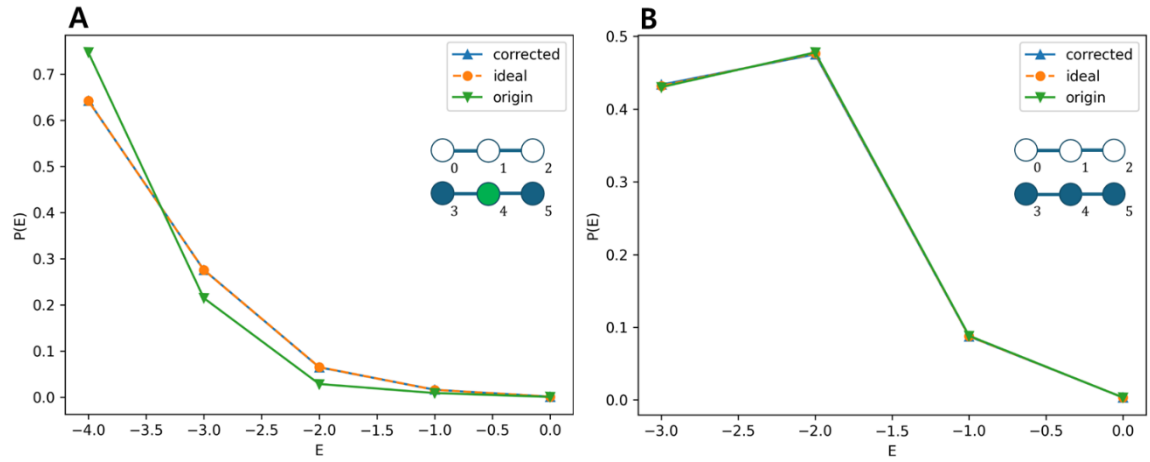


Figure 1. $P(E)$ represent frequencies of states with total energy E during all simulation steps. For the move set frequencies, only Move_Rot and Move_Local are set to 1, and all others are 0. If all energy scales are identical, there is no significant discrepancy between original version and corrected version (B). However, if chain have various energy scales, there is a difference (A).

3. Errors in calculating Rosenbluth weight factor for an array of beads

Certain MC moves, such as MV_Snake and MV_Trans, change multiple rotational bonds simultaneously. Initially, all rotational bonds are dissociated, and an array of participating beads is constructed. The rotational states of each bead are determined sequentially along the array. The rotational bond is exclusive, so once a bond is formed, the bonded partner is excluded from the potential candidates for subsequent beads in the array. The original LaSSI did not handle this hierarchical dependency properly, leading to a break in detailed balance.

3.1. Workflow in original version

The same procedure used for calculating the Rosenbluth weight factor of a single bead was applied to an array of beads. However, directly reusing this approach caused improper calculations of the backward Rosenbluth weight factor. To satisfy microscopic reversibility, the backward transition should be executed in the same way as the forward transition. Therefore, even if a neighboring bead has a bond partner, if it has a later order in the array, it should be considered a candidate (Figure 2). The original LaSSI excluded it, causing an error.

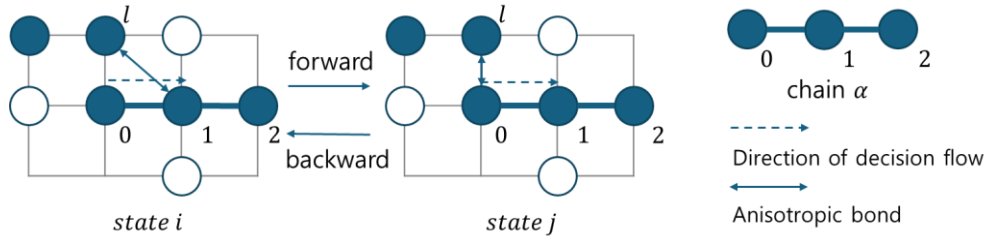


Figure 2. At the first step in the backward transition, all rotational bonds are dissociated. Bead l should be connected with bead 1 after a bond selection for bead 0 through sequential decisions. Therefore, when deciding an rotational bonding partner for bead 0, bead l should be considered a candidate.

3.2. Workflow in corrected version

To handle the hierarchical dependency properly, the search for potential rotational bond partners for bead a , which has index ' i ' in the array, was changed as follows:

1. List all 26 adjacent beads to bead ' a '.
2. Check the beads in the adjacent list sequentially. If a bead has a bond partner with an index ' j ' where $j < i$, exclude it.

3.3. Impact on the result energy distribution

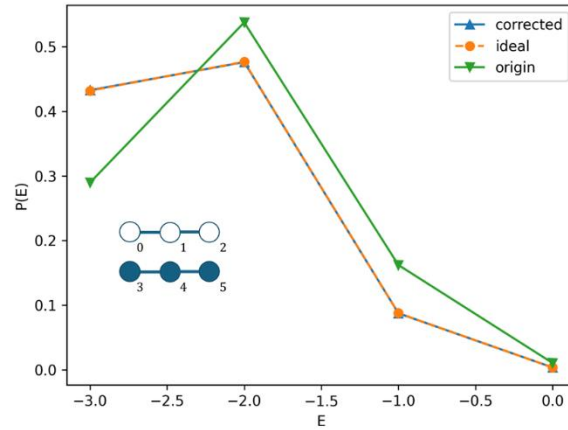


Figure 3. $P(E)$ represent frequencies of states with total energy E during all simulation steps. Frequencies for MV_STROT, MV_LOCAL, MV_MTLOCAL, MV_SNAKE, MV_TRANS are set to 1, and all others are set to 0. There is a significant discrepancy between original version and corrected version.