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New implementations of replica-exchange method for simulations of complex systems: Designed-walk and deterministic replica-exchange methods

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Abstract

Two new methods of replica-exchange method (REM) are tested for a two-dimensional Ising spin model. The first method is the deterministic replica-exchange method (DETREM) which uses a differential equation based on Gibbs sampling method instead of Metropolis criteria. The other is the designed-walk replica-exchange method (DEWREM) which determines the trajectory of replica in temperature space without random walk. This method gives more number of tunneling events than conventional REM, where the tunneling event is a round-trip of temperature from the lowest to the highest back to the lowest. We examined physical quantities such as magnetization and susceptibility. Our new methods reproduced the results of the conventional random-walk REM.

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Keywords: replica-exchange method; differential equation; detailed balance condition

Introduction

Replica-exchange method (REM) is a popular way to accelerate configurational sampling. In temperature REM, the replica-exchange process is based on the Metropolis criterion (Hukushima and Nemoto (1996)) or a conditional probability in Gibbs-sampling (Chodera and Shirts (2011)). To evaluate performance and condition for convergence is important to analyze the method. For analysis of performance, many works were performed to increase the efficiency of REM such as by optimized replica allocation, mathematical analysis, and replica exchange attempt frequency. We recently proposed the deterministic replica-exchange method (DETREM) (Urano and Okamoto (2014)) which controls replica exchange using a new variable and its evolution of a differential equation. Moreover, to analyze the performance of REM, some measurements were proposed. A simple measurement of this method is tunneling events (Berg and Neuhaus (1992); Mitsutake et al. (2003b)), which is the number of round-trip between the low-

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est temperature and highest temperature. This measurement shows us whether replicas visit many temperatures in temperature space. Hence, increase of tunneling events leads to improvement of REM efficiency. We also proposed the designed-walk REM (DEWREM) (Urano and Okamoto (2015)) in which replicas follow a pre-planned route of replica exchange in temperature space instead of a random walk. In this article, we first introduce the methods of DETREM and DEWREM and, then, results are presented by these new methods for a two-dimensional Ising model system.

Methods

We now give the details of our methods. We prepare M non-interacting replicas at M different temperatures. Let the label i (=1, $\cdots$, M) stand for the replica index and label m (=1, $\cdots$, M) for the temperature index. We represent the state of the entire system of M replicas by $X = \left\{x^{[1]}_{m(1)}, \cdots, x^{[M]}_{m(M)}\right\}$, where $x^{[i]}_m = \left\{q^{[i]}, p^{[i]}\right\}_m$ are the set of coordinates $q^{[i]}$ and momenta $p^{[i]}$ of particles in replica i (at temperature T_m). The probability weight factor for state X is given by a product of Boltzmann factors: $W_{\rm REM}(X) = \prod_{i=1}^{M} \exp\left[-\beta_{m(i)} H(q^{[i]}, p^{[i]})\right]$, where $\beta_m (= 1/k_{\rm B}T_m)$ is the inverse temperature and $H(q, p)$ is the Hamiltonian of the system. We consider exchanging a pair of replicas i and j corresponding to temperatures T_m and T_n, respectively: $X = \left\{\cdots, x^{[i]}_m, \cdots, x^{[j]}_n, \cdots\right\} \rightarrow X' = \left\{\cdots, x^{[j]\prime}_m, \cdots, x^{[i]\prime}_n, \cdots\right\}$, where $x^{[i]\prime}_n \equiv \left\{q^{[i]}, p^{[i]\prime}\right\}_n$, $x^{[j]\prime}_m \equiv \left\{q^{[j]}, p^{[j]\prime}\right\}_m$, and $p^{[j]\prime} = \sqrt{\frac{T_m}{T_n}} p^{[j]}$, $p^{[i]\prime} = \sqrt{\frac{T_n}{T_m}} p^{[i]}$ (Sugita and Okamoto (1999)).

Here, the transition probability $\omega(X \rightarrow X')$ of Metropolis criterion for replica exchange is given by

$$\omega(X \rightarrow X') = \min\left(1, \frac{W_{\rm REM}(X')}{W_{\rm REM}(X)}\right) = \min(1, \exp(-\Delta)), \tag{1}$$

where

$$\Delta = \Delta_{m,n} = (\beta_n - \beta_m)(E(q^{[i]}) - E(q^{[j]})). \tag{2}$$

Because each replica visits various temperatures followed by the transition probability of Metropolis algorithm, REM performs a random walk in temperature space.

We now review two new REMs, which are based on random walks in temperature space. Without loss of generality, we can assume that M is an even integer and that $T_1 < T_2 < \cdots < T_M$. The conventional REM (Hukushima and Nemoto (1996); Sugita and Okamoto (1999)) is performed by repeating the following two steps:

1. We perform a conventional MD or MC simulation of replica i $(= 1, \cdots, M)$ at temperature T_m $(m = 1, \cdots, M)$ simultaneously and independently for short steps.
2. Pairs of exchange attempts are selected in replica pairs with neighboring temperatures, for example, for the odd pairs (T_1, T_2), (T_3, T_4),$\cdots$, (T_{M-1}, T_M) or even pairs (T_2, T_3), (T_4, T_5),$\cdots$, (T_{M-2}, T_{M-1}).

All the replica pairs thus selected are attempted to be exchanged according to the Metropolis transition probability in Eqs. (1) and (2) with $n = m + 1$. We repeat Steps 1 and 2 above until the end of the simulation. The canonical ensemble at any temperature is reconstructed by reweighting techniques (Ferrenberg and Swendsen (1989); Mitsutake et al. (2003a)).

We next present the deterministic replica-exchange method (DETREM) (for a general formalism, see, Urano and Okamoto (2014)). Only Step 2 is different from the conventional REM. At first, we introduce an internal state $y_{m,n}$ as an index of a pair of replicas i and j at temperatures T_m and T_n, and consider the following differential equation:

$$\frac{dy_{m,m+1}}{dt} = \sigma_m \frac{1}{1 + \exp(\Delta_{m,m+1})}, \tag{3}$$

where t is a virtual time, $\Delta_{m,m+1}$ is the same as in Eq. (2) with $n = m+1$, and the signature σ_m of the pair of (T_m, T_{m+1}) changes to 1 or -1 to control the signature of the change of y_m which monotonically increases or decreases. In Step 2, instead of applying the Metropolis criterion in Eqs. (1) and (2), we solve the differential equation in Eq. (3) for the internal states $y_{m,m+1} \in \{-1, 1\}$ for (T_m, T_{m+1}), where the total number of internal states is M-1 with the following pairs: (1,2), (2,3), $\cdots$, (M-1,M) for the random-walk DETREM and the pairs: (1,2), (3,4), $\cdots$, (M-1,M) and (2,3), (4,5), $\cdots$, (M-2,M-1) for designed-walk REM. The replica exchange is done as follows (Urano and Okamoto (2014)):

$$\text{if updated } y_{m,m+1} \gtreqless \pm 1, \text{ then } (T_m, T_{m+1}) \rightarrow (T_{m+1}, T_m),\ y_{m,m+1} \leftarrow y_{m,m+1} \mp 1, \sigma_m \leftarrow \mp 1.$$

For the random-walk DETREM, if $y_{m,m+1}$ performs exchanges, $y_{m+1,m+2}$ is not time evolved and $y_{m+2,m+3}$ is evolved to avoid the leap exchange of temperature such as from T_m to T_{m+2}.

We then explain the algorithm of designed-walk replica-exchange method (DEWREM) briefly (for detail,s see. (Urano and Okamoto (2015)). The designed temperature walk can be implemented to both conventional Metropolis REM and DETREM (and other REMs) as follows. Namely, DEWREM is performed by repeating the following steps.

1. We perform a conventional MD or MC simulation of replica i $(= 1, \cdots, M)$ at temperature T_m $(m = 1, \cdots, M)$ simultaneously and independently for short steps.

2. Replica exchange is attempted for all the odd pairs (T_1,T_2), (T_3,T_4), $\cdots$, (T_{M-1},T_M).

3. Repeat Steps 1 and 2 until all odd pairs perform replica exchange exactly once. Namely, once a pair is exchanged, the exchanged pair stops exchange attempts and keep performing the simulation in Step 1 with the new temperatures. Replica exchange attempt in Step 2 is repeated until all the other odd pairs finish exchanges.

4–6. Repeat Steps 1–3 where the odd pairs in Steps 2 and 3 are now replaced by the even pairs (T_2,T_3), (T_4,T_5), $\cdots$, (T_{M-2},T_{M-1}).

7. The cycle of Steps 1 to 6 is repeated until the number of cycles is M, which is equal to the tunneling count and all replicas have the initial temperatures.

8. Begin the above cycle of Steps 1–7 with Steps 1 to 3 and Steps 4 to 6 interchanged.

These eight steps are repeated until the end of the simulation. This algorithm satisfies the detailed balance condition because the wait for remaining replica-exchange pairs is complemented by the symmetric swap in Step 8.

Simulation conditions

In order to test the effectiveness of the present methods, we performed simulations with conventional REM, DETREM and DEWREM for a 2-dimensional Ising model. The lattice size in a square lattice was 128 (hence, the number of spins was $N = 128^2 = 16384$). We have performed conventional random-walk simulation and designed-walk simulation of both Metropolis REM and DETREM. We have also performed a mixed random-walk and designed-walk simulation of DETREM, where we repeated the two walks alternately. The total number of replicas M was 40 and the temperatures were 1.50, 1.55, 1.60, 1.65, 1.70, 1.75, 1.80, 1.85, 1.90, 1.94, 1.98, 2.01, 2.04, 2.07, 2.10, 2.13, 2.16, 2.19, 2.22, 2.25, 2.28, 2.31, 2.34, 2.358, 2.368, 2.38, 2.40, 2.42, 2.44, 2.47, 2.51, 2.57, 2.63 ,2.69, 2.75, 2.82 ,2.90, 3.00, 3.10, and 3.15. Boltzmann constant k_B and coupling constant J were set to 1. Thus, $\beta = 1/k_BT = 1/T = \beta^*$, and the potential energy is given by $E(\mathbf{s}) = -\sum_{<i,j>} s_i s_j$, where $s_i = \pm 1$, and the summation is taken over all the nearest-neighbor pairs in the square lattice.

For the conventional random-walk REM and DETREM, replica-exchange attempt was made every 1 MC step. One MC step here consists of one Metropolis update of spins. The total number of MC steps for all the simulations was 100,000,000. To integrate Eq. (3), we used the fourth-order Runge-Kutta method with virtual time step $dt = 1$. For DEWREM, replica-exchange attempt was made every 20, 50, 100, 150, 200 MC steps in the conventional REM simulations and every 20, 50, 100, 150, 200, and 250 MC steps in the DETREM simulations (see Table 1). The mixed-walk simulation was performed in which after $4M(= 160)$ even-odd or odd-even cycles of designed-walk simulations (replica-exchange attempt was made at every 20 MC steps) were performed, 200,000 MC steps (which roughly corresponds to $2M$ cycles) of random-walk simulations (replica-exchange attempt was made at every 1 MC step) were performed, and then this procedure was repeated. For reweighting analyses (Ferrenberg and Swendsen (1989); Mitsutake et al. (2003a); Shirts and Chodera (2008)), the total of 10,000 spin state data were taken with a fixed interval of 1,000 MC steps at each temperature from the REM simulations.

Results

Table 1 lists the mean tunneling counts per replica for each method, which is the number of times where the replicas visit from the lowest temperature through the highest temperature and back to the lowest during the simulation. The

Table 1. The mean number of tunneling counts per replica.

TC	Random walk		Designed walk								Mixed walk
	Met	DETREM	Met				DETREM				DETREM
Interval	1	1	20	50	100	150	20	100	150	200	1 & 20
Mean	173	178	292	197	131	99	231	93	69	55	293
± SD	9.5	8.8	56	41	27	21	48	20	15	11	6

TC, Interval, SD, Met stand for tunneling counts, the number of MC steps between replica-exchange attempts, standard deviation, and REM based on Metropolis criterion, respectively. The frequency (1 & 20) of Mixed walk means that it was 1 MC step for random walk REM and 20 MC steps for designed-walk REM.

mean tunneling counts per replica of the designed-walk simulations at every 10 MC attempts were about twice larger. These large tunneling counts imply that in designed-walk method all replicas traversed more efficiently in temperature space, and our design to maximize the tunneling counts for all replicas without random walks was successful. For the mixed-walk simulation, the maximum tunneling count was about twice larger than that of random-walk DETREM. The mean tunneling count was almost the same as that of designed-walk DETREM.

We next examine physical quantities obtained from the designed-walk simulations with various replica-exchange attempt frequencies and mixed walk simulations and compare them to those from the conventional random-walk simulations (for average energy density and heat capacity, see, Urano and Okamoto (2015)).

Fig. 1(a) and Fig. 1(b) show magnetization M as a function of T obtained from the random-walk and designed-walk simulations of Metropolis REM and DETREM, for DETREM including the mixed-walk simulation. They were obtained by the reweighting techniques. However, in the DEWREM simulations, the magnetization with high frequency replica-exchange attempts at $T = 2.25$ near the exact critical temperature $T_c = 2.269$ are deviated slightly compared to the results of the random-walk simulation. The slow relaxation at lower temperature than the exact critical temperature is also confirmed in susceptibility as shown in Fig. 2. Fig. 2(a) and Fig. 2(b) show susceptibility χ as a function of temperature obtained from the random-walk and designed-walk simulations of Metropolis REM and DETREM, for DETREM including the mixed-walk simulation. This figure shows that by extending intervals of replica-exchange attempts DEWREM simulation can reproduce the results of random-walk REM in both conventional REM and DETREM. Moreover, these physical quantities show that repeating a random walk and designed walk simulation in mixed-walk simulation is an efficient way to increase the accuracy of results and the number of tunneling counts at the same time.

Conclusions

In this article, we introduced DETREM and DEWREM . The new methods reproduced the results of conventional REM simulation. For DETREM, this means that the analysis of internal states and its evolution give a new method to analyze the REM. For DEWREM, replica-exchange trials with artificial order cause a correlation between replicas which have no direct interactions among replicas. However, by reducing the correlation with a mixed walk, longer interval replica-exchange trials, and combination of non-neighboring replica-exchange pairs, DEWREM will be more efficient.

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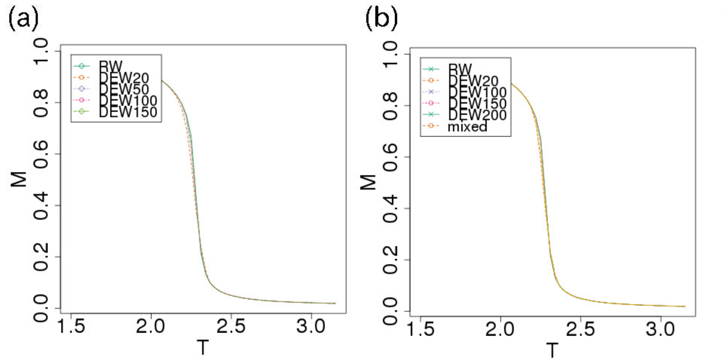

Fig. 1. Magnetization M as a function of T from the (a) REM, (b) DETREM simulations including the mixed-walk simulation. The error bars are smaller than the symbols.

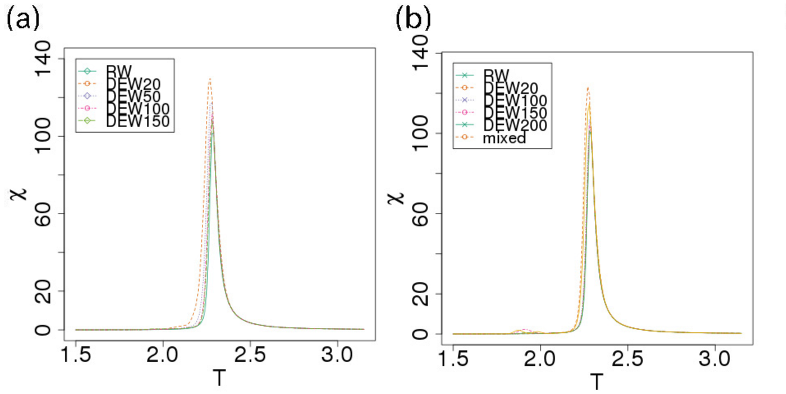

Fig. 2. Susceptibility χ as a function of T from the (a) REM, (b) DETREM simulations including the mixed-walk simulation. The error bars are smaller than the symbols.

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References

Berg, B.A., Neuhaus, T., 1992. Multicanonical ensemble: A new approach to simulate first-order phase transitions. Phys. Rev. Lett. 68, 9.

Chodera, J.D., Shirts, M.R., 2011. Replica exchange and expanded ensemble simulations as gibbs sampling: Simple improvements for enhanced mixing. J. Chem. Phys. 135, 194110.

Ferrenberg, A.M., Swendsen, R.H., 1989. Optimized monte carlo data analysis. Phys. Rev. Lett. 63, 1195–1198.

Hukushima, K., Nemoto, K., 1996. Exchange monte carlo method and application to spin glass simulations. J. Phys. Soc. Jpn. 65, 1604–1608.

Mitsutake, A., Sugita, Y., Okamoto, Y., 2003a. Replica-exchange multicanonical and multicanonical replica-exchange monte carlo simulations of peptides. i. formulation and benchmark test. J. Chem. Phys. 118, 6664.

Mitsutake, A., Sugita, Y., Okamoto, Y., 2003b. Replica-exchange multicanonical and multicanonical replica-exchange monte carlo simulations of peptides. ii. application to a more complex system. J. Chem. Phys. 118, 6676–6688.

Shirts, M.R., Chodera, J.D., 2008. Statistically optimal analysis of samples from multiple equilibrium states. J. Chem. Phys. 129, 124105.

Sugita, Y., Okamoto, Y., 1999. Replica-exchange molecular dynamics method for protein folding. Chem. Phys. Lett. 314, 141 – 151. doi:10.1016/S0009-2614(99)01123-9.

Urano, R., Okamoto, Y., 2014. Deterministic replica-exchange method without pseudo random numbers for simulations of complex systems, submitted for publication. e-print arXiv:1412.6959.

Urano, R., Okamoto, Y., 2015. Designed-walk replica-exchange method for simulations of complex systems, submitted for publication. e-print arXiv:1501.00772 .