Parallel Random Tunneling Algorithm for Structural Optimization of Lennard-Jones Clusters up to $N = 330$

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A random tunneling algorithm (RTA) is derived from the terminal repeller unconstrained subenergy tunneling (TRUST) algorithm, and the parallelization of the RTA is implemented with an island parallel paradigm. Combined with the techniques of angular moving, the parallel random tunneling algorithm (PRTA) is applied to the optimization of Lennard-Jones (LJ) atomic clusters, and all the global minima of LJ clusters with size up to 200 are successfully located. For the optimization of larger cluster, a PRTA with an improved seeding technique is developed and successfully applied to the optimization of LJ151−LJ309. Furthermore, the optimized structures of LJ309−330 with the PRTA, which have not been studied before, are also provided.

1. INTRODUCTION

In theoretical chemistry, to achieve a deep understanding of clusters and reveal the evolution properties with cluster size, lots of work has been done on the study of the global optimal structures by combining theoretical optimization and experimental results, whereas the task of minimizing the energy of cluster is notoriously difficult because the number of local minima tends to grow exponentially with cluster size N.[1] Consequently, it is still a great challenge to develop an efficient algorithm for the structural optimization of clusters.

As a benchmark system for the optimization algorithms in this field, the Lennard Jones (LJ) atomic cluster has been investigated intensively, and the minimum energies and structures of LJ clusters with $N \leq 309$ are available on the Internet.[2,3] Many methods have been applied to the optimization of LJ clusters. Incorporating particular physical insights into the LJ problem, most global minima of the LJ clusters for $N \leq 309$ are found by the lattice based methods.[4,5] In the lattice method, the search is performed on the specific structural lattices chosen by previous knowledge, whereas unbiased methods that make no assumptions regarding cluster geometry attract more interest because these methods have good generalization and can be applied to other cluster systems. The methods include genetic algorithm (GA),[6−9] simulated annealing (SA),[10−14] Basin-Hopping,[1,15] and others.[16−18] An innovative GA approach was proposed by Deaven and Ho (DH-GA)[6,7] and made great progress for the structural optimization of cluster systems. In DH-GA, the genetic operators run directly on the configuration space of clusters. DH-GA finds most of the global minima of LJ clusters up to $N = 100$ but fails for some nonicosahedral clusters, e.g. LJ38, LJ75−77 and LJ102−104. Hartke improves the DH-GA with the concept of niches to treat the hard cases and finds all the minima of LJ clusters up to 150 atoms.[9] Basin-Hopping, in which the Monte Carlo algorithm is combined with a Basin-Hopping potential energy surface (PES) deformation technique, is proposed by Wales et al. and has been successfully applied to the optimization of LJ clusters up to $N = 110$.[15,19] Moreover, the tetrahedral structure of LJ98 is also found with the Basin-Hopping method.[20] The Self-Consistent Basin-to-Deformed-Basin Mapping (SCBDBM) can locate the minima of LJ clusters with the size up to 100 except for LJ75, LJ76 and LJ77.[16] PFAEA is a parallel algorithm that employs a very fast annealing technique and evolutionary population strategy and can find all the minima of LJ clusters up to 116 atoms.[14] The optimization of a larger cluster is very difficult for the unbiased algorithm. Only several methods have been employed to investigate the clusters with a size greater than 150, and the optimizations are only performed on several clusters. For example, the minima of LJ200 and LJ300 are first reported by Xue.[12] The minima of LJ192 and LJ201 are first reported by Wales.[15] The new minima of LJ185 and LJ186 are found by Hartke's phenotype GA,[3] whereas there is still no global optimization method that can reach the size of 309. Consequently, it is a great challenge to break the limitation for a global optimization method.

In this study, a parallel random tunneling algorithm (PRTA) was developed and applied to the structural optimization of LJ clusters with a size lower than 330. The random tunneling algorithm (RTA) is derived from the terminal repeller unconstrained subenergy tunneling (TRUST) algorithm.[18] TRUST is a deterministic global optimization approach that has been successfully used in exploratory seismology.[21−23] Based on the subenergy tunneling technique of TRUST, the stochastic algorithm RTA was developed for the purpose of structural optimization in our former work.[18] The serial RTA has successfully located the known minima of LJ clusters containing up to 100 atoms.[18] To improve the efficiency, PRTA is run in parallel with the island paradigm,[24] and an improved seeding technique is developed. All the known minima of LJ clusters with the size lower than 309 were successfully located by PRTA with the improved seeding technique. Furthermore, with the PRTA, the optimal structures of clusters from LJ310 to 330 are also investigated, and the best structures obtained in the optimization are provided.

2. METHOD

2.1. Random Tunneling Algorithm. A population of configurations is adopted in RTA to explore the solution

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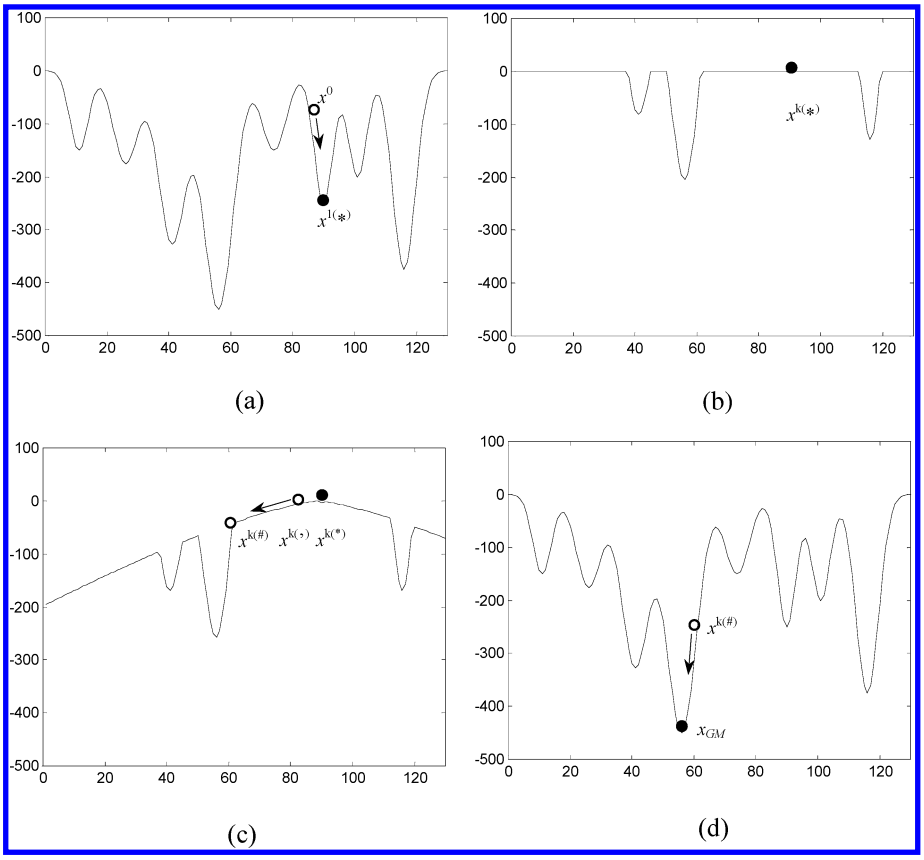

Figure 1. The schematic diagram of the random tunneling algorithm (RTA). (a) Initialization. The line represents the initial objective function. (b) Random tunneling begin, and subenergy transformation is performed. (c) Random tunneling on the virtual function landscape. (d) Local minimization is the global minimum.

space.[18] A schematic diagram of RTA is demonstrated in Figure 1. In the initialization, each individual is generated randomly and is then minimized to the local minimum with the limited memory BFGS method (L-BFGS)[23,25] as shown in Figure 1(a). After initialization, the random tunneling and local minimization are executed circularly, and $\boldsymbol{x}^k$ indicates the individual at the kth round. First, the subenergy transformation is performed on the energy landscape of each individual to convert all points lying high above current local minimum approximately to zero, see Figure 1(b). Second, a penalty function is applied to transform current minimum to local maximum, see Figure 1(c). Equation 1 is the virtual objective function combined the subenergy transformation (first term) and terminal repeller effects (second term)

$$E(\boldsymbol{x},\boldsymbol{x}^*) = \log\,(1/[1 + \exp(-(\hat{f}(\boldsymbol{x}) + \beta))]) - (3/4)\rho(\boldsymbol{x} - \boldsymbol{x}^*)^{4/3}\theta(\hat{f}(\boldsymbol{x})) \quad (1)$$

where β and ρ are the positive parameters, with the values 2 and 20 in the study, respectively. $\hat{f}(\boldsymbol{x}) = f(\boldsymbol{x}) - f(\boldsymbol{x}^*)$, and $\boldsymbol{x}^*$ is the current local minimum. θ is a Heaviside function. Third, the individual $\boldsymbol{x}^k$ is perturbed to $\boldsymbol{x}^{k(\prime)}$. Starting with $\boldsymbol{x}^{k(\prime)}$, the dynamic system will flow down on the transformed energy landscape $E(x,x^*)$ until $\boldsymbol{x}^{k(\#)}$ is found, at which the value of the Heaviside function is changed to zero. The local phase is started after the random tunneling, see Figure 1(d). If $\boldsymbol{x}^{k(\#)}$ is found, then optimize $\boldsymbol{x}^{k(\#)}$ to the next minimum $\boldsymbol{x}^{k+1(*)}$. Repeat the random tunneling procedure until the global minimum is found or the count k reaches a set value.

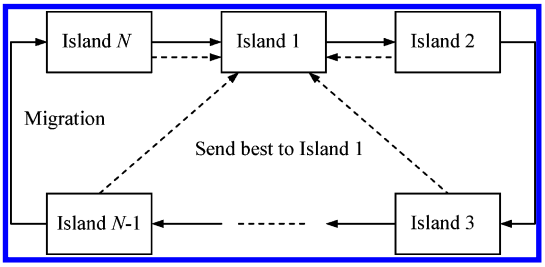

Figure 2. The ring topology of island parallel paradigm employed in PRTA.

In the optimization of LJ clusters, the technique of angular moving[14,18,26,27] is adopted and performed at the end of RTA for the further optimization of the best cluster.

2.2. Parallelization. To improve the efficiency of RTA, a coarse-grained island model with a ring topology (see Figure 2) is adopted to implement the PRTA. In PRTA, the population is divided to N_p (number of processors) subpopulations, and each subpopulation is allocated to a processor. In the optimization, N_p is 8, and the size of the whole population is 240.

The island model is one of the successful coarse-grained paradigm of the parallel genetic algorithms.[24,28,29] In the island model, the population is divided into smaller subpopulations which are optimized independently and simultaneously. Therefore, the isolated subpopulation evolves as on an island. Migration is an important operator in an island model. Periodic migrations of some selected individuals between islands allow for injecting new diversity into subpopulations. The role of migration is to exchange information from one island to another. It has been shown

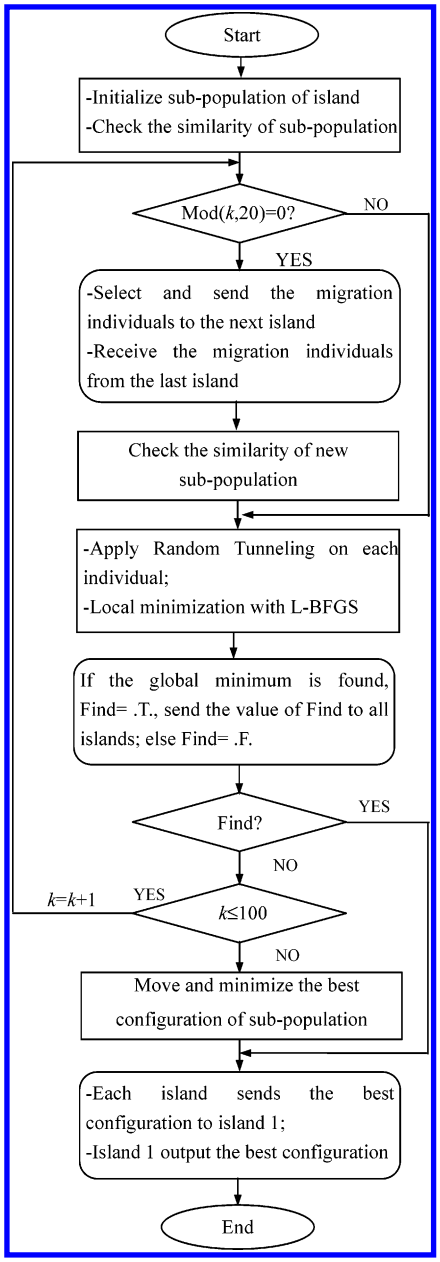

Figure 3. Flowchart of PRTA.

that it is equivalent to the GA crossover operator at the abstraction level of the population. Topology defines the spatial structure of connections between subpopulations.[29] The ring topology is easy to be implemented and is mostly used in an island parallel model.[28]

The migration operator has two parameters, the migration rate (i.e., the quantity of individuals that migrate at once) and migration frequency.[24,29] The two parameters determine the amount of information exchanged between islands. The communication time increase with the two parameters. Furthermore, too large of a migration rate and high frequency will inevitably lead to the high similarity of the whole population and preconvergence of the optimization. In general, the migration rate is in the range of 10−20%. In the current study, empirical parameters are adopted. The migration rate is 20%, and the migration interval is 20 circles.

The flowchart of PRTA is given in Figure 3. Starting with a randomly generated or seeded subpopulation, a serial RTA is run on each island. After 20 circles of the random tunneling and the local minimization, the migration operator is performed. Twenty percent of the total subpopulation with comparatively lower energy is selected as the migrants, and then the migrants are sent from island N-1 to the next island (e.g. island N) along with the direction indicated by the arrows with the solid line in Figure 2. After the migrants are received, the subpopulations will be reconstructed by selecting the individuals with lower energies from the migrants and the old subpopulation.

In the optimization of LJ clusters with $N \leq 309$, all nodes will check if the known global minimum is found after each circle of the random tunneling and local minimization, as indicated in Figure 3. The node 1 (e.g. island 1 shown in Figure 2) is responsible for gathering the values of the boolean variable Find from all nodes. If one node has found the global minimum, node 1 will send the true value to the Find of all other nodes. Therefore, the optimization will stop if the global minimum is found on one node.

When the parallel random tunneling algorithm is finished, the angular moving of outside atoms is performed on the best configuration of each subpopulation. Finally, the first processor (island) will gather the best individual of each island as the arrows with dashed lines are shown in Figure 3 and select the configuration with the lowest energy as the optimal result.

In the present work, PRTA program is performed on a cluster of workstations (COW). The cluster is a distributed memory computer system containing eight computers with two Pentium III 1G CPUs each. Parallel code is written according to the Message Passing Interface (MPI) library and the standard MPI software used is MPICH-1.2.1.

2.3. An Improved Seeding Technique. The seeding technique is very helpful for the structural optimization of clusters and has been employed in many optimization works of clusters.[15,8,26,27] Most of the seeds which have been used before are the clusters with one atom less or one atom more or with several atoms less than the cluster needs to be optimized. The effect of seeding depends on whether the seeds have similar optimal configurations with the current cluster, whereas it is difficult to choose an appropriate seed, because we often have no knowledge about the structure of the cluster that needs to be optimized. Therefore, a universal seeding strategy will be more practical.

From the optimization results up to now, the most important structural motif in LJ clusters is icosahedron, and the other important configuration is Marks' decahedron.[2,3] Therefore, two motif seeds, a completed icosahedron and a completed Marks' decahedron, are added besides the seed of LJN-1 to develop a more universal seeding technique. Wolf and Landman have employed the seeds of structural motif including icosahedron, decahedron and octahedron, and the seeding technique is effective in finding the minima with N up to 100.[8]

In the improved seeding technique, the population is initialized from the three different seeds: LJN-1, a completed icosahedron, and a completed Marks' decahedron. In addition, the ratio of individuals seeded from the three seeds is set as 1:2:2. First, the coordinates of the M atoms are read from the seed file directly, and the remaining $N-M$ atoms are placed outside of the seed. N and M represent the size of the cluster to be optimized and the number of atoms in seed, respectively. The spherical coordinates of the $N-M$

Table 1. Efficiency Analysis of the PRTA with Island Paradigm[c]

number of processors	size of subpopulation	N_m[a]	T_{total}[b] (s)	speedup
1	240		11164.26	
2	120	24	5567.38	2.01
4	60	12	2809.86	3.97
6	40	8	1867.02	5.98
8	30	6	1405.57	7.94
12	20	4	943.30	11.84
16	15	3	709.07	15.74

[a] Number of migrating individuals on each island. [b] Total run time, averaged from five runs. [c] Population size: 240, the migration ratio: 20%, the total iterations of random tunneling process: 100, and the cluster size: LJ98.

atoms are calculated as follows: (1) Coordinates of the first atom are generated randomly at the outside of the seed. (2) Taking the first atom as the center atom, an angular range is calculated in proportion to the value of *N*-*M*. Other atoms are restricted to generate in the range. (3) Randomly generate a new atom in the range. (4) Check the distances between the atoms generated in the seeding. If one atom is too near to one of the other atoms, the program will decide to accept this atom or not according to a probability, and the probability of acceptance is 20%. If accept the current atom, repeat (3) and generate the next atom; otherwise repeat (3) and regenerate the coordinates of the atom. With the seeding technique, the initial configurations are obtained and are optimized to the local minima using L-BFGS.

In the following procedures, the coordinates of atoms in the seed are fixed, and only parameters of the new generated atoms are optimized. The similarity checking is implemented on each kind of population initialized from different seed respectively, and only the newly generated coordinates are considered for calculating similarity. In the migration, the migrants are selected from the three kinds of clusters whose initial configurations are initialized from the three different seeds. After 100 generations of random tunneling, the atoms at the outside of the cluster are further optimized with angular moving, and the angular displacement data sets are made from the outside atoms of the motif seeds. After that, another 100 generations of random tunneling will be started if the global minimum has not been found, and all parameters will be optimized.

3. RESULTS AND DISCUSSION

3.1. Efficiency of PRTA. The speedup of PRTA with a different number of processors is given in Table 1. In evaluating the performance of the island parallel model, the time for angular moving is not included. It is shown that the parallel model adopted in PRTA is efficient because a near liner speedup is obtained, and the time for communication takes less than 0.5% of the total run time. The speedup behaves as a small decrease when the number of processors is larger than 8. In the optimization of LJ clusters, the PRTA is run on 8 processors and the total population is 240, and the performance of PRTA is satisfying.

The success ratios of the three hard cases LJ38, LJ75 and LJ98 are also investigated, and they are found to be 10 out of 10, 2 out 100, and 6 out of 50 runs, respectively. The success ratios of PRTA for the three hard cases are better than those of PFAEA,[14] but no significant improvement can be found when PRTA was compared with the serial RTA.[18] Additionally, two migration strategies in the island model have been investigated. The one we used is selecting the best individuals for the migration and replacing the worst individuals of the subpopulation with the immigrants. In another method, both the selection of migration individuals and the replacement are at random. Test results show that the two methods have no remarkable difference in the optimization ability and computational time.

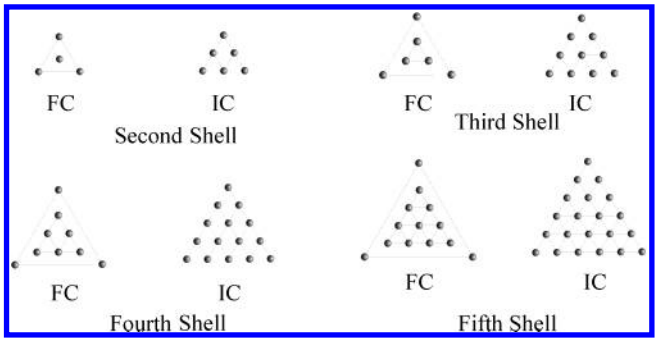

Figure 4. Comparison of IC and FC in the configuration of one face of icosahedron.

3.2. The Optimization Results of PRTA. The minima of LJ clusters given in ref 2 and 3 with *N* lower than 309 were reproduced successfully by the PRTA. Except for LJ206 and LJ223 with energies of -1271.663247ϵ and -1389.687271ϵ, respectively, which are slightly lower than the reported energies of -1271.663226ϵ and -1389.687270ϵ,[3] other minima are the same as the reported values with the accuracy reaching the sixth decimal place.

To test the optimization ability of PRTA, LJ3−150 and several larger clusters including LJ_{160}, LJ_{170}, LJ_{180}, LJ_{190} and LJ_{200} were optimized with the initial configurations generated randomly. All the known minima of these clusters are successfully reproduced. It indicates that the optimization ability of pure PRTA can reach 200 atoms. For the clusters of larger sizes 151−309, the improved seeding technique was employed to improve the efficiency.

Generally, if the cluster to be optimized has *N* atoms, the seed was chosen from the optimal structure of the cluster with *N*-1 or *N*+1 atoms,[15,26,27] whereas the seeding technique is not always effective, because there are several cases that may result in fault. Sometimes the neighboring LJ clusters have different configurations such as icosahedron and Marks' decahedron, e.g. the structural motif of the two neighboring clusters of LJ187 and LJ188 are icosahedron and Marks' decahedron, respectively. Consequently, the optimal decahedral structure of LJ188 cannot be found by seeding from icosahedral LJ187. Furthermore, for the clusters with icosahedral motif, the outside atoms of the LJ*N* and LJ*N*-1 may have a different packing style, e.g. FC and IC.[4] The difference of FC and IC is depicted in Figure 4. The transformation widely exists in the LJ clusters, for example, the neighboring clusters of LJ153(FC) and LJ154(IC), LJ168(FC) and LJ169-(IC), LJ176(IC) and LJ177(FC) etc. Even for the clusters with the same structural motif and packing style, the configurations of the outside atoms are not always similar. For example, the IC configurations of LJ202 and LJ203 are quite different, if seeding from LJ202, the energy of LJ203 is -1250.080278ϵ, whereas the lowest energy of LJ203 is -1250.106610ϵ. Therefore, the seeding technique by adding

Table 2. Optimization of LJ151−309 by PRTA with Seeding Technique

N	seed[a]	motif[b]	*N*	seed[a]	motif[b]	*N*	seed[a]	motif[b]
151−153	1	FC	191−192	1	MD	245	2	IC
154	2	IC	193	2	IC	246−259	1	IC
155	2	FC	194−199	1	IC	260	2	IC
156−159	1	FC	200	0	IC	261	1	IC
160	0	FC	201−202	1	IC	262	2	IC
161−168	1	FC	203	2	IC	263	1	IC
169	0	IC	204−216	1	IC	264	2	IC
170	0	IC	217	2	IC	265−271	1	IC
171−176	1	IC	218−219	1	IC	272	2	IC
177	2	FC	220	2	IC	273−282	1	IC
178	1	FC	221−232	1	IC	283	2	IC
179	1	IC	233	2	IC	284	1	IC
180	0	IC	234−235	1	IC	285	2	IC
181−187	1	IC	236	3	MD	286−289	1	IC
188	3	MD	237−238	1	MD	290	2	IC
189	1	MD	239	2	IC	291−309	1	IC
190	0	MD	240−244	1	IC			

[a] In the optimization with seeding technique, three kinds of seeds are used simultaneously. The number given is the successful seed: 1, seed of LJ*N*-1; 2, complete icosahedron LJ147; 3, Marks' decahedron. 0 indicates seeding technique is not used. [b] Structural motifs in LJ clusters. IC: Having icosahedral core with outside atoms packed in IC style; FC: Icosahedral core with outside atoms packed in FC style; MD: Marks' decahedron.

one atom or removing one atom from the seed is far from enough. By adding more general motif seeds along with the seeding strategy for the outside atoms, the improved seeding technique is tested in the study.

In the range of 151−309, LJ clusters were optimized mainly by PRTA with the improved seeding technique. In the first 100 random tunneling circles of RTA with seeding, only parameters of the newly generated atoms are optimized. The parameters of problem are greatly reduced, and the optimization process is very efficient. Generally, only the clusters with an incomplete icosahedral core, e.g. LJ203, need to be optimized further without fixing any parameters. The details about the successful seed, which lead to the optimal structures, are tabulated in Table 2. The icosahedral motif seed is the completed icosahedron LJ147, and the Marks' decahedral seeds for LJ188 and LJ236 are LJ102 and LJ192, respectively. In the table, the number of the successful seed 2 or 3, i.e. the motif seed of icosahedron or decahedron, indicate there are changes in configuration at the sizes. All the minima of the special cases mentioned above are found successfully with initial configuration seeding from motif seeds. Consequently, the improved seeding technique is effective and more universal than the previous seeding technique for the structural optimization of LJ clusters.

Furthermore, it is notable that the generation strategy of the outside atoms, in which new atoms are generated in a restricted range, is important and helpful. The outside atoms always tend to be compact because the cluster will have a larger number of nearest neighbor contacts than a loose packing style. In the work, it is found that if the new atoms are generated randomly at the outside of the seed, the efficiency of the optimization is much worse than the seeding method with restriction.

3.3. The Optimized Structures of LJ310−330. With the help of the improved seeding technique, PRTA are also employed to investigate the configurations of LJ310−330. The energies and structures, provided in Table 3 and Figure 5 respectively, are the best results from 100 runs of PRTA. To the best of our knowledge, these LJ clusters from LJ310 to LJ330 have not been investigated before.

Table 3. Lowest Energies of LJ310−330 Obtained by PRTA with Seeding along with the First Finite Difference of Energy ΔE, the Number of Nearest Neighbors N_{nn} and Its First Finite Difference ΔN_{nn}

N	*E*	motif	ΔE	N_{nn}	ΔN_{nn}
310	−2012.098565	FC	−4.879565	1551	3
311	−2017.838110	IC	−5.739545	1555	4
312	−2024.652123	IC	−6.814013	1560	5
313	−2031.396713	IC	−6.744590	1565	5
314	−2038.173131	IC	−6.776418	1570	5
315	−2044.980920	IC	−6.807789	1575	5
316	−2052.140774	FC	−7.159854	1581	6
317	−2058.468188	FC	−6.327414	1586	5
318	−2064.797071	FC	−6.328883	1591	5
319	−2071.874428	IC (2)[a]	−7.077357	1593	2
320	−2079.108659	IC (1)	−7.234231	1599	6
321	−2086.343410	IC	−7.234751	1605	6
322	−2093.169666	IC	−6.826256	1610	5
323	−2099.995344	IC	−6.825678	1615	5
324	−2106.682396	FC (1)	−6.687052	1621	6
325	−2113.917005	FC	−7.234609	1627	6
326	−2120.916334	FC (1)	−6.999329	1632	5
327	−2128.150926	FC	−7.234592	1638	6
328	−2134.488722	FC	−6.337796	1643	5
329	−2141.430727	IC	−6.942005	1645	2
330	−2148.373452	IC (1)	−6.942725	1650	5

[a] Number given in parentheses is the number of missing atoms in the icosahedral core.

In Figure 5, the background lattice represents the completed icosahedron core LJ309, and the outside atoms are plotted with balls that are connected by a stick. LJ319, LJ320, LJ324, LJ326 and LJ330 have an incomplete icosahedral core, and the locations of the missing atoms are also given as the vacancies of the background icosahedral lattice in Figure 5.

The structural growth property of LJ clusters has the characteristic to form the next completed icosahedron from a smaller icosahedron. The magic numbers with a completed icosahedron of medium size are 55, 147, 309, 561, and it is assumed that the processes to forming the outside icosahedral lattices of different sizes have similarity. Consequently, the structural properties of LJ310−330 can be compared with the known mimima of LJ56−72 and LJ148−160. The

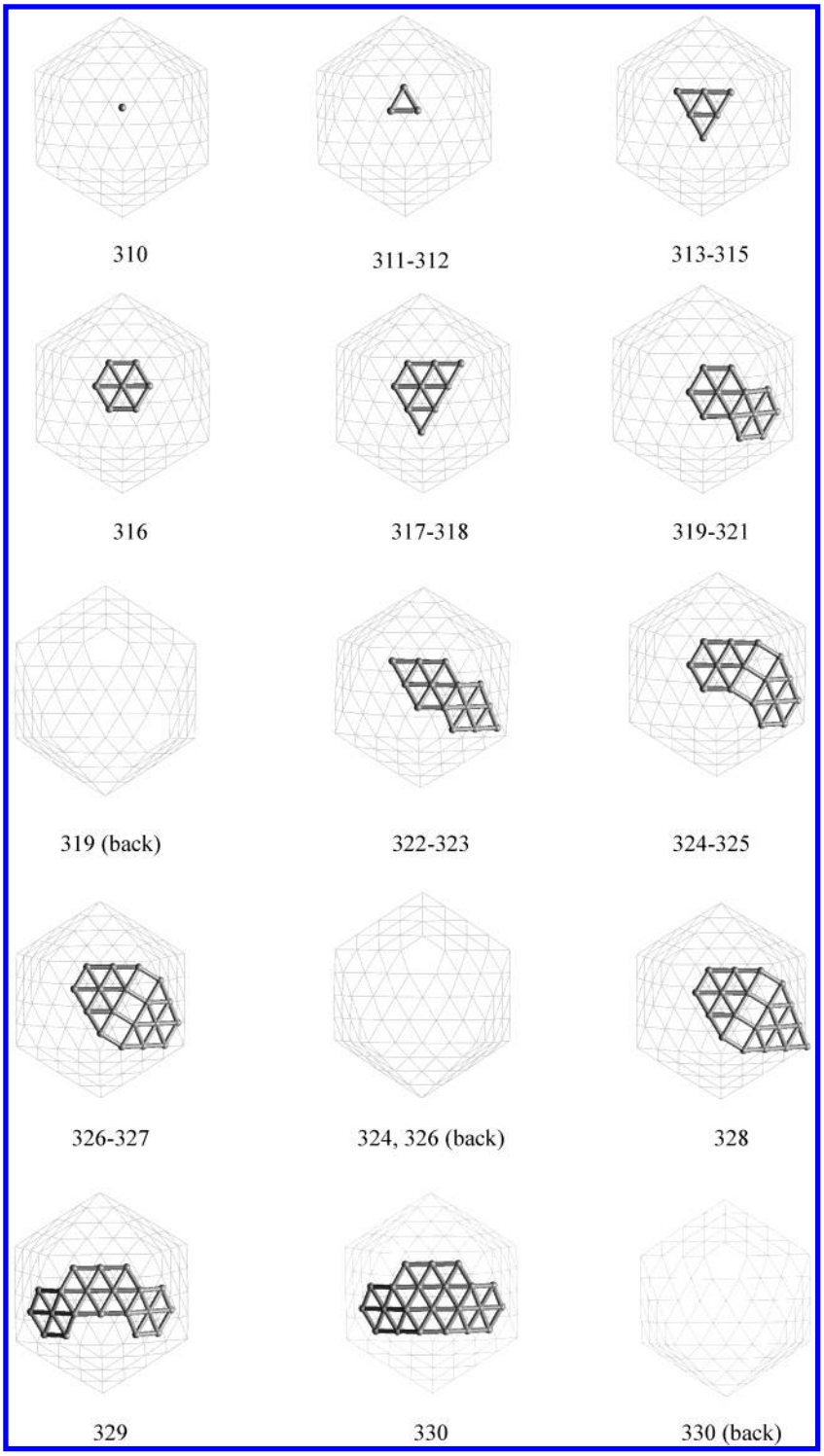

Figure 5. Structures of LJ310−LJ330.

variation of the first finite differences of energy ΔE and the number of nearest neighbor contacts ΔN_{nn} from LJ50 to LJ330 are plotted in Figure 6. In Figure 6(a), by comparing the ΔN_{nn} and ΔE of LJ310−330 with those clusters of LJ56−72 and LJ148−160, it is found that the ΔN_{nn} and ΔE of LJ310−330 are in a normal range. Furthermore, the structures of LJ310−330 are compared with those of LJ148−160. At the beginning of the packing for the new outside layer, FC and IC configurations are competing for the optimal structures. It is indicated that the structural transitions from LJ315 (IC) to LJ316 (FC), from LJ318 (FC) to LJ319 (IC), and from LJ323 (IC) to LJ324 (FC) are quite similar to the transitions from LJ150 (IC) to LJ151 (FC), from LJ153 (FC) to LJ154 (IC), and from LJ154 (IC) to LJ155 (FC),[5] respectively. Therefore, the energy and structures of clusters LJ310−330 are reasonable and may be good candidates for the optimal configurations.

4. CONCLUSION

The parallel random tunneling algorithm was proposed and successfully located all the known minima of LJ clusters up to $N = 309$. With a randomly initialized population, the

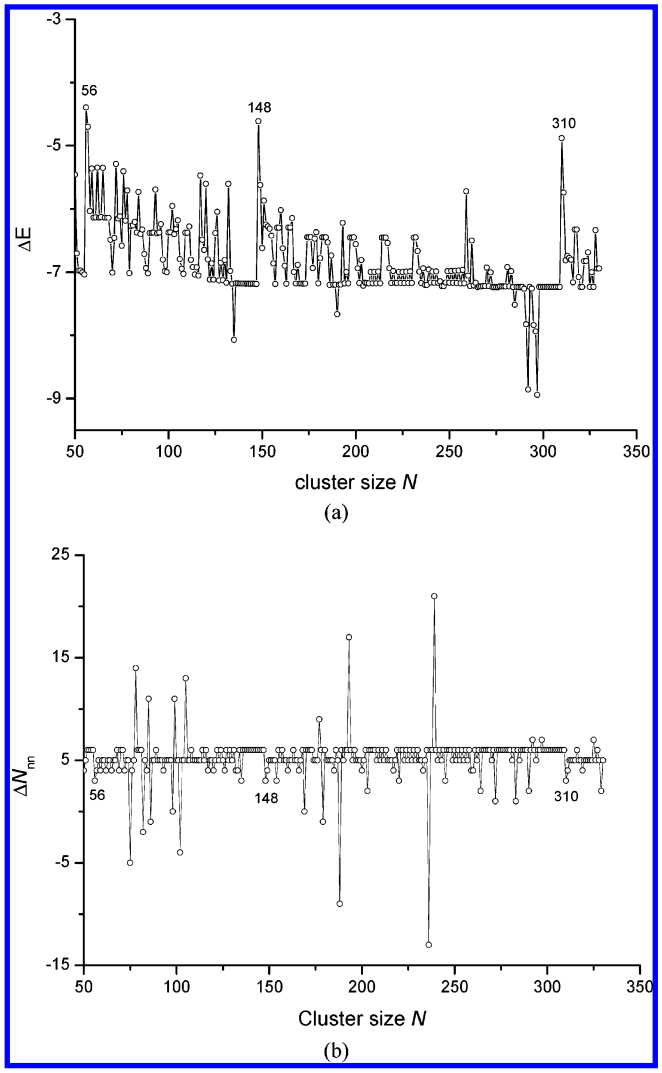

Figure 6. Feasibility of the minima of LJ310−330. (a) The first finite differences of energy ΔE versus cluster size N. (b) The first finite difference of the number of nearest neighbor contacts ΔN_{nn} versus cluster size N.

maximal size that PRTA can reach is LJ200. By adding two motif seeds, an icosahedon and a Marks' decahedron, the improved seeding technique was successfully applied to the optimization of the larger cluster size $151 \leq N \leq 330$. By comparing the optimization results and analyzing the structures of LJ clusters, it is shown that the improved seeding technique is reasonable and efficient for the structural optimization of cluster. Consequently, the optimization ability of PRTA is good, and the improved seeding technique may be helpful for the studies of other clusters by changing the motif seeds.

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REFERENCES AND NOTES

(1) Wales, D. J.; Scheraga, H. A. Global Optimization of Clusters, Crystals and Biomolecules. *Science* **1999**, *285*, 1368−1372.
(2) Wales, D. J.; Doye, J. P. K.; Dullweber, A.; Naumkin, F. Y. The Cambridge Cluster Database. http://brian.ch.cam.ac.uk.
(3) LJ Clusters with Minimum Potential for $148 \leq N \leq 309$. http://www.vcl.uh.edu/~cbarron/LJ_cluster/LJpottable.html.
(4) Northby, J. A. Structure and Binding of Lennard-Jones Clusters: $13 \leq N \leq 147$. *J. Chem. Phys.* **1987**, *87*, 6166−6177.
(5) Romero, D.; Barrón, C.; Gómez, S. The Optimal Geometry of Lennard-Jones Clusters: 148−309. *Comput. Phys. Commun.* **1999**, *123*, 87−96.
(6) Deaven, D. M.; Ho, K. M. Molecular Geometry Optimization with a Genetic Algorithm. *Phys. Rev. Lett.* **1995**, *75*, 288−291.
(7) Deaven, D. M.; Tit, N.; Morris, J. R.; Ho, K. M. Structural Optimization of Lennard-Jones Clusters by a Genetic Algorithm. *Chem. Phys. Lett*. **1996**, *256*, 195−200.
(8) Wolf, M. D.; Landman, U. Genetic Algorithms for Structural Cluster Optimization *J. Phys. Chem. A* **1998**, *102*, 6129−6137.
(9) Hartke, B. Global Cluster Geometry Optimization by a Phenotype Algorithm with Niches: Location of Elusive Minima, and Low-Order Scaling with Cluster Size. *J. Comput. Chem.* **1999**, *20*, 1752−1759.
(10) Wille, L. T. Minimum-energy Configuration of Atomic Clusters: New Results Obtained by Simulated Annealing. *Chem. Phys. Lett.* **1987**, *133*, 405−410.
(11) Ma, J.; Straub, J. E. Simulated Annealing using the Classical Density Distribution. *J. Chem. Phys.* **1994**, *101*, 533−541.
(12) Xue, G. Molecular Conformation on the CM-5 by Parallel Two-Level Simulated Annealing. *J. Glob. Opt.* **1994**, *4*, 187−208.
(13) Schelstraete, S.; Verschelde, H. Finding Minimum-Energy Configurations of Lennard-Jones Clusters using an Effect Potential. *J. Phys. Chem. A* **1997**, *101*, 310−315.
(14) Cai, W. S.; Jiang, H. Y.; Shao, X. G. Global Optimization of Lennard-Jones Clusters by a Parallel Fast Annealing Evolutionary Algorithm. *J. Chem. Inf. Comput. Sci.* **2002**, *42*, 1099−1103.
(15) Wales, D. J.; Doye J. P. K. Global Optimization by Basin-Hopping and the Lowest Energy Structures of Lennard-Jones Clusters Containing up to 110 Atoms. *J. Phys. Chem. A* **1997**, *101*, 5111−5116.
(16) Pillardy, J.; Liwo, A.; Scheraga, H. A. An Efficient Deformation-Based Global Optimization Method (Self-Consistent Basin-to-Deformed-Basin Mapping (SCBDBM)). Application to Lennard-Jones Atomic Clusters. *J. Phys. Chem. A* **1999**, *103*, 9370−9377.
(17) Curotto, E.; Freeman, D. L.; Doll, J. D. A J-Walking Algorithm for Microcanonical Simulations: Applications to Lennard-Jones Clusters. *J. Chem. Phys.* **1998**, *109*, 1643−1647.
(18) Jiang, H. Y.; Cai, W. S.; Shao, X. G. A Random Tunneling Algorithm for Structural Optimization Problem. *Phys. Chem. Chem. Phys.* **2002**, *4*, 4782−4788.
(19) Doye, J. P. K.; Wales, D. J.; Miller, M. A. Thermodynamics and the global optimization of Lennard-Jones clusters. *J. Chem. Phys.* **1998,** *109*, 8143−8153.
(20) Leary, R. H., Doye J. P. K. Tetrahedral Global Minimum for the 98-atom Lennard-Jones cluster. *Phys. Rev. E* **1999**, *60*, R6320−R6322.
(21) Barhen, J.; Protopopescu, V.; Reister, D. TRUST: A Deterministic Algorithm for Global Optimization. *Science* **1997**, *276*, 1094−1097.
(22) Cetin, B. C.; Barhen, J.; Burdick, J. W. Terminal Repeller Unconstrained Subenergy Tunneling (TRUST) for Fast Global Optimization. *J. Optimiz. Theory App.* **1993**, *77*, 97−126.
(23) Liu, D. C.; Nocedal, J. On the Limited Memory BFGS Method for Large Scale Optimization. *Math. Progm.* **1989**, *45***,** 503−528.
(24) Cantù-Paz, E. *Efficient and Accurate Parallel Genetic Algorithms*; Kluwer Academic Publishers: 2000.
(25) Betts, S.; Browne, S.; Dongarra, J.; Grosse, E.; McMahan, P.; Rowan, T. Netlib Repository, A Collection of Mathematical Software, Papers, and Databases. http://www.netlib.org.
(26) Cai, W. S.; Feng, Y.; Shao, X. G.; Pan, Z. X. Optimization of Lennard-Jones Atomic Clusters. *J. Mol. Struct.* (*Theochem*) **2002**, *579*, 229−234.
(27) Cai, W. S.; Feng, Y.; Shao, X. G.; Pan, Z. X. Global Optimization of $(C60)_N$ Molecular Clusters. *Chem. Phys. Lett.* **2002**, *359*, 27−34.
(28) Wagner, D.; Calegari, P.; Pizarosso, M. Parallel island-based genetic algorithm for radio network design. *J. Parallel Distributed Comput.* **1997**, *47*, 86−90.
(29) Cantù-Paz, E. A summary of research on parallel genetic algorithms. Illinois Genetic Algorithms Laboratory, Report No. 95007, 1995.

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