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An energy-based perturbation and a taboo strategy for improving the searching ability of stochastic structural optimization methods

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Abstract

An energy-based perturbation and a new idea of taboo strategy are proposed for structural optimization and applied in a benchmark problem, i.e., the optimization of Lennard–Jones (LJ) clusters. It is proved that the energy-based perturbation is much better than the traditional random perturbation both in convergence speed and searching ability when it is combined with a simple greedy method. By tabooing the most wide-spread funnel instead of the visited solutions, the hit rate of other funnels can be significantly improved. Global minima of (LJ) clusters up to 200 atoms are found with high efficiency.

1. Introduction

Determination of the global minimum of Lennard–Jones (LJ) clusters by numerical global optimization methods has been intensively studied [1–3]. LJ clusters consist of identical atoms interacting by the LJ potential:

$$V(r) = 4\varepsilon \sum_{i<j} \left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6} \right], \quad (1)$$

where r_{ij} represents the distance between atoms i and j, and the reduced units are generally used, i.e., $\varepsilon = \sigma = 1$. The LJ potential is not only interesting as a model for noble gases but also serves as a popular benchmark for evaluating new algorithms for structural optimization. Most of the known global minima for small LJ clusters are based on icosahedral packing. The exceptions, global minima of LJ_{38} (fcc like truncated octahedron), $LJ_{75–77}$, $LJ_{102–104}$ (Marks decahedron) and LJ_{98} (Leary tetrahedron [4]) serve as particularly interesting cases. At these magic numbers, the global minima lie in a very deep funnel on the LJ potential energy surface, while the lowest-energy icosahedral structure acts as a trap in a much wider funnel.

How to explore new solutions based on current solutions is very important for the convergence speed and searching ability in a stochastic optimization method, such as genetic algorithm (GA) [5,6], basin-hopping [1,7], adaptive immune optimization algorithm [8,9], dynamic lattice searching (DLS) [10], etc. In most stochastic structural optimization methods, the perturbation (or 'mutation' in GA) on a structure is to randomly move some atoms in the structure, and the same probability for all the atoms in a structure is generally used. In fact, in real systems, atoms with higher potential energy certainly have higher reactivity at a certain temperature. This means that atoms with higher potential energy should have a higher chance to be moved in the optimization.

Taboo search (TS) is a search procedure for solving complex combinatorial optimization problems of a general type proposed by Glover [11]. This procedure has also been extended to optimization of continuous-valued functions [12]. The basic idea of this method is

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to locally and repeatedly modify a solution while memorizing these modifications to avoid visiting the same solutions twice or in a cyclic manner. Modifications are stored in the taboo list that forbids their use for a certain number of iterations. Although TS has been successfully applied to the structural optimization of atomic clusters [13–15], the calculation results do not show obvious improvement compared to some other stochastic global optimization methods. The reason may be that, because there are too many parameters and the searching space is continuous, it is too difficult to taboo so many visited solutions. Furthermore, how to decide the taboo area around the visited solution is another problem because it is difficult to measure the similarity of two structures.

Previous studies on the potential energy landscape (PEL) have shown that the PEL can be characterized as being composed as various funnels [16,17]. From a randomly generated structure, it is easy to step into the bottom of the funnel containing the structure, while the transition between different funnels is generally difficult. For some cases, such as some magic numbers of the LJ clusters, the local funnel is much wider, while the global funnel is much deeper. Therefore, for a stochastic method, it may be entrapped into the wide-spread funnel easily, and hence the other funnels (including the global one) will have little chance to be visited. The basic idea of our new taboo strategy is that the local funnel bottom will be tabooed instead of the visited solutions. By this approach, other funnels which containing the conformations with different packing style to the tabooed funnel will have higher chances of being located.

2. Simple greedy method with energy-based perturbation

To test the performance of the proposed energy-based perturbation, a simple greedy method (SGM) is adopted. Similar to the basin-hopping method, the newly generated structures and the structures after perturbation will be minimized immediately by using a local minimization procedure (the quasi-Newton L_BFGS [18] routine is used in this study), and all the operations are based on the local minima. SGM is somewhat similar to monotonic sequence basin-hopping (MSBH) [19]. The SGM procedure is rather simple. It starts with a randomly generated starting local minimum X_0, then repeatedly explores new solutions around X_k ($k = 0, 1, 2,\ldots$) by perturbations, i.e., if a structure with lower potential energy than X_k is found, accept it as the next starting local minimum X_{k+1}, until no structure with potential energy lower than X_k can be found within N_{try} perturbations (where N_{try} is a constant).

In stochastic optimization methods, the convergence speed and searching ability are strongly related to the strategy of how to explore new solutions based on current solutions. In real systems, atoms with higher potential energy are more reactive at a certain temperature. Inspired by this fact, the energy-based perturbation is proposed, i.e., atoms with higher potential energy will have a higher chance to be selected to move. In fact, similar idea have been adopted in a number of approaches to cluster optimization. In [5], only the single atom with highest potential energy is mutated. In [20], the atoms on the outer shell, which have higher potential energy, are frequently moved. In this study, a full study on the energy-based perturbation will be carried out.

For a given minimized structure, the potential energy of each atom in the structure is calculated at first. Then, the selection probability of atom i is

$$P(i) = \exp(V_i/T)/\sum_{j=1}^{N}\exp(V_j/T),$$

where V_i is the potential energy of atom i, N is the cluster size, and T is the temperature (in this study, $T = 0.5$ is used). The number of atoms to be moved is

$$N_{\text{mov}} = 1 + 0.1 \times N \times r^3,$$

where r is a random number between 0 and 1. If an atom is selected, it will be moved to a random place in the cluster. In [9], the selected atoms are randomly moved to the surface of the cluster. But, by chance, we found that moving the selected atom to the center of the cluster is more suitable. In this study, the atom is moved to a random location with distance $R = R_0 \times r^2$ to the center of mass of the cluster, where r is a random number between 0 and 1, and $R_0 = 1 + (3N/4\pi\sqrt{2})^{1/3}$ [21].

To enhance the efficiency of exploring new solutions around a given local minimum X, a small population is used (the maximal population size used in this study is $N_{\text{pop}} = 5$). At the start of the exploration, the population size is 1 with the starting local minimum, then perform the following circulation: (1) Randomly select a structure from the population and perform energy-based perturbation described above on the structure to obtain a new minimized structure Y. (2) If Y is better than X, the exploration succeeds and will be terminated with Y as the result. (3) Otherwise, if the population size is less than N_{pop}, put the structure into the population and increase the population size by 1. (4) If the population size is equal to N_{pop} and Y is better than the worst one in the population, replace it. (5) If the number of perturbations is smaller than N_{try}, go to step (1). (6) Otherwise, the starting local minimum X is regarded as a funnel bottom on the PEL and this exploration will be terminated. The population strategy may make the transitions between similar conformations easier compared to the exploration based on only one single local minimum.

To verify the performance of the proposed strategies, comparisons of the convergence performance between SGM with random perturbation (R-SGM), SGM with energy-based perturbation but without population

(E-SGM), and SGM with energy-based perturbation and with a small population size $N_{pop} = 5$ (EP-SGM) are carried out for the case of the optimization of LJ_{75}. R-SGM and MSBH have the same basic frame, but are different in the strategy of perturbation. The perturbation strategy of R-SGM is similar to that of E-SGM. The only difference is that, in R-SGM, the atoms to be moved are randomly selected. Fig. 1 shows a comparison of the three methods. The numbers of funnel bottoms located from 1000 independent SGM runs for R-SGM (Fig. 1a), E-SGM (Fig. 1b), EP-SGM (Fig. 1c) are 158, 112, and 33, respectively. This indicates that the energy-based perturbation and a small population can significantly decrease the number of funnel bottoms, which means they can increase the searching ability. By comparison of the convergence speed, i.e., the average number of local minimizations per SGM run (excluding the final fixed number N_{try}), the numbers for the three methods are 711, 211 and 258, respectively. This shows the energy-based perturbation can greatly increase the convergence speed. Furthermore, compared to MSBH [19], the convergence speed of EP-SGM is also higher, for example, the number of local minimizations required for convergence in MSBH for LJ_{30}, LJ_{80}, and LJ_{110} are 286, 844, and 1256, respectively, while in EP-SGM, the values are only 88, 214, and 593, respectively.

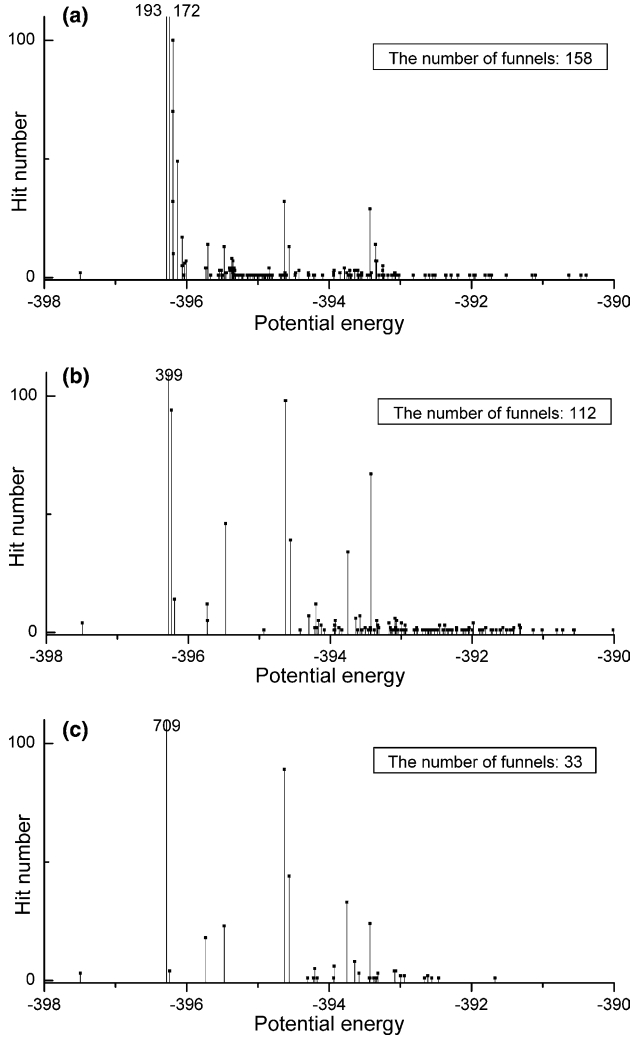

Fig. 1. Convengence performance for the case of the optimization of LJ_{75} based on 1000 independent runs (termination condition is $N_{try} = 500$). (a) SGM with random perturbation (R-SGM); (b) SGM with energy-based perturbation but without population (E-SGM); (c) SGM with energy-based perturbation and with a small population $N_{pop} = 5$ (EP-SGM). The number of funnels means the number of different results located by SGM.

Results of MSBH and EP-SGM for some selected cases are given in Table 1. It can be seen that, for small cases ($N < 70$), EP-SGM does not offer many advantages compared to MSBH. But for the large cases, EP-SGM is much better than MSBH both in the number of located funnel bottoms and the number of local minimizations per hit of global minimum. For small cases, the two methods have similar hit rate of the global minimum, which means that the searching ability is enough for convergence in both methods. But with increasing cluster size, the hit rate of MSBH decreases much more than that of EP-SGM, which means that the searching ability of MSBH decreases more rapidly than that of EP-SGM. Therefore, the energy-based perturbation can increase the searching ability especially for clusters of larger size. For larger cluster sizes, e.g., $N \geqslant 160$, the number of located funnel bottoms is too large to make the hit rate of the global minimum small due to the exponential increase of the searching space. Maybe, for large cluster size, evolutionary strategies are more suitable than the simple greedy strategy. The reason for choosing the simple greedy strategy in this study is to show the performance of the energy-based perturbation more clearly. The basic idea of the energy-based perturbation can certainly be adopted in other stochastic structural optimization methods, such as genetic algorithm, simulated annealing algorithm, basin-hopping method, and Monte-Carlo method.

3. SGM with a taboo strategy

Although the energy-based perturbation can increase the convergence speed and searching ability, e.g., as shown in Fig. 1 for the LJ_{75}, the hit rate of the icosahedral funnel bottom increases greatly when the population strategy and energy-based perturbation is adopted. However, there is no obvious increase of the hit number of the global minimum for non-icosahedral packing. Therefore, a taboo strategy is proposed with the basic idea that, if the wide-spread icosahedral funnel is tabooed, the hit rate for other packing (including the global one) should increase.

To determine the area to be tabooed, the connectivity table (CT) [9] is used for cluster similarity checking. The distance of two local minima v and w can be calculated by

Table 1
Performance of the MSBH and EP-SGM algorithms in terms of number of located funnel bottoms, average number of local minimizations (LM) required to find the global minimum (excluding the final fixed overhead of N_{try}), and global minimum hit rate over 1000 independent runs

N	MSBH			EP-SGM		
	Funnels	LM	Hit rate	Funnels	LM	Hit rate
30	2	739	0.382	3	132	0.662
38[a]	3	2875	0.124	7	1739	0.144
40	9	279	0.849	6	175	0.898
50	7	460	0.868	7	264	0.796
60	12	388	0.948	5	255	0.800
70	90	1526	0.630	35	428	0.797
75[a]	75	152 000	0.004	33	86 000	0.003
80	132	2009	0.420	54	317	0.671
90	198	4699	0.206	88	2218	0.271
98[a]	210	180 000	0.006	100	27 200	0.019
100	221	9128	0.122	104	2525	0.211
102[a]	245	36 028	0.031	116	13 880	0.041
110	288	40420	0.031	192	3951	0.150
120				310	9666	0.077
130				351	5880	0.134
140				328	2243	0.350
150				369	3230	0.224
160				476	10 263	0.083
170				650	38 578	0.025
180				729	36 946	0.028
190[a]				719	358 195	0.003
200				746	56 550	0.019

For EP-SGM, $N_{try} = 500$ at $N \leqslant 80$ and $N_{try} = 1000$ for the others.
[a] Global minimum with non-icosahedral packing.

$$D(v,w) = \left(1 + \left|n_{nn}^{v} - n_{nn}^{w}\right|\right)^{1/2} \sum_{i=1}^{M} i \times \left|\mathrm{CT}^{v}(i) - \mathrm{CT}^{w}(i)\right|, \quad (2)$$

where n_{nn} is the number of nearest neighbor contacts, CT(i) means the number of atoms whose coordination number is i in the cluster, and M is the maximal coordination number for an atom (for general cases M is 12, but for disorder packing M may be larger than 12). Considering that the funnel bottom generally lies in the center of a funnel, structures with distances less than the taboo radius, R_T, are thought to be contained in the tabooed funnel and will be discarded directly.

To test the performance of the taboo strategy, the optimization of LJ_{75} is also selected as a test case. Fig. 1c has shown that the most dominant conformation of LJ_{75} is the anti-icosahedron (FC), and hence it is selected as the tabooed funnel. Fig. 2 shows the hit number of structures with other packing by increasing R_T from 0 (no taboo) to 1100 with an increment of 100. As we expected, hit number for other packing motifs obviously increases with the R_T. For small R_T (100–400), the hit number of IC increases most rapidly, while the hit numbers of other packings do not increase significantly. The reason may be that IC is most similar to FC and they are both local bottoms in the icosahedral funnel, and hence the ones that should drop into the FC bottom will turn to the IC bottom when the FC is tabooed. But with R_T increasing, the hit number of IC decreases and falls to 0 at $R_T \geqslant 600$ because the whole icosahedral funnel is tabooed, and hence the hit numbers of other packing increase obviously especially for TE (the 75-atom TE can be a fragment of 98-atom TE). In fact, the hit number of the global minimum (ID) also increases greatly, e.g., the hit number of ID increases from 3–4 to about 20–24 with an increase of about 5–8 times, which is similar to that of TE although the latter seems to increase more rapidly in the figure.

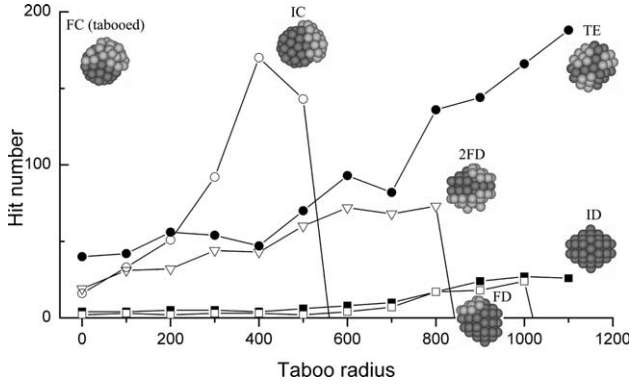

Fig. 2. Hit numbers of various packings as a function of the taboo radius (R_T) based on 1000 independent runs. FC is icosahedron with anti-layers; IC is icoshhedron; ID is Marks decahedron; FD is decahedron with anti-layers; 2FD is decahedron with anti-layers on both sides; TE is Leary tetrahedron.

In constrast to the results of some other stochastic methods, the TE funnel seems not to be narrow, which

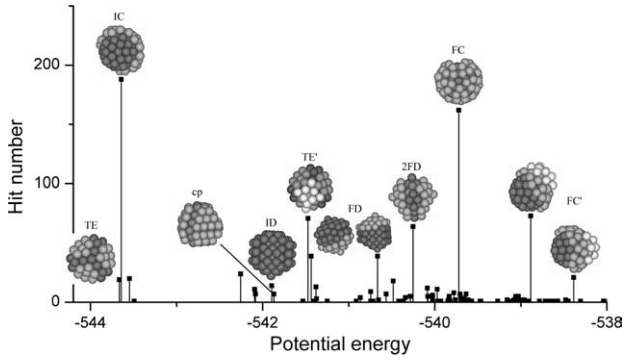

Fig. 3. Conformational analysis by counting the hit number of various funnel bottoms for the case of LJ_{98} based on 1000 independent runs. cp is close packed; TE′ is similar to TE but with some different atom locations (white ones); FC′ is similar to FC but with outer layers on the anti-layers and more disordered.

agrees with the result of the dynamic lattice searching method (DLS) [10]. To further study this phenomenon, a conformational analysis for LJ_{98} is given in Fig. 3 by counting the hit numbers of all located funnel bottoms out of 1000 runs. It can be seen that the icosahedral funnel (including IC, FC, and FC′) is certainly the most dominant one. The tetrahedral funnel (including TE and TE′) and decahedral funnel (including ID, FD, and 2FD) are not too narrow, while the close packed funnel (including cp and fcc) can only act as the background in the figure. The result is significantly different from the small cases, e.g., LJ_{75} (Fig. 1c) and LJ_{38}. The reason may be that for small clusters the transition between funnels is too easy, and hence the SGM results may differ greatly from the real PEL. As shown in this figure, the decahedral funnel contains various metastable bottoms (ID, FD, and 2FD), and the total hit number of the decahedral funnel is not small, which means the decahedral funnel is not too narrow. Despite this, the hit number of a certain bottom may be small, which is determined by both potential energy and entropy [22] (for entropy 2FD > FD > ID). The tetrahedral funnel is also similar to the decahedral funnel (for entropy TE′ > TE).

4. Conclusions

An energy-based perturbation and a taboo strategy were proposed for the structural optimization problem. With the benchmark of Lennard–Jones clusters, the energy-based perturbation was proved to be much better than the traditional random perturbation both in convergence speed and searching ability using a simple greedy method, and by tabooing the most wide-spread funnel, other funnels can be visited with a higher probability. Both the energy-based perturbation and the taboo strategy should be used universally in stochastic methods for structural optimization.

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