

Ab Initio Parallel Replica Molecular Dynamics Method

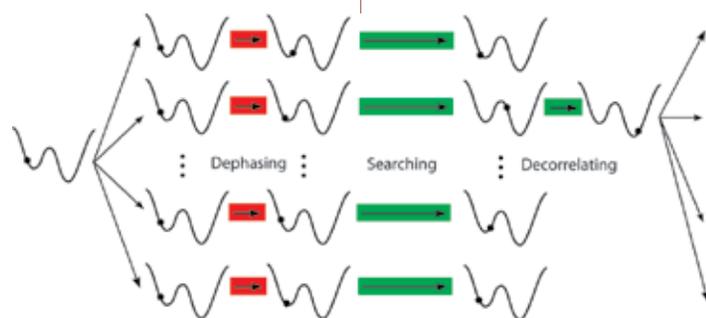
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Dynamical behavior of infrequent-event systems is characterized by a trajectory that resides in a potential basin for many vibrational periods before undergoing a transition to a new basin. Modeling dynamics of the infrequent-event systems is very challenging, since very large time scales need to be considered in order to capture their relevant behavior. One of the ways to tackle this challenge is through the use of accelerated molecular dynamics methods, such as parallel replica molecular dynamics (PRMD) [1].

The PRMD method parallelizes the molecular dynamics (MD) simulation in the time domain. The only assumption made is that the infrequent events studied obey the first-order kinetics. The PRMD method is illustrated in Fig. 1. The system studied is first prepared in an initial state, which is then replicated into M copies sent to M processors. All copies (or replicas of the system) are then dephased in order to eliminate correlations between them. After the dephasing stage, each processor carries out an independent MD simulation, exploring the phase space in the original basin

M times faster than a single MD trajectory would. Once the transition is detected, the simulation clock is advanced by the sum of the accumulated MD time in each replica. The trajectory that made the transition is allowed to continue for an additional amount of

Fig. 1. Schematic of the PRMD method. Total simulation time is a sum of MD time spent searching or decorrelating (green arrows). Dephasing time (red arrows) does not advance the simulation clock.



time called decorrelation time, during which recrossing or other correlated events may occur. The simulation clock is then advanced by decorrelation time, the system in the new state is again replicated into M copies, and the whole process is restarted.

To obtain forces acting on atoms in each step of an MD simulation, current implementations of the PRMD method rely on the use of empirical potentials fitted to empirical data or electronic structure calculations. Accurate empirical potentials are hard to develop and they must be redetermined if one changes an atom type. Therefore, ideally, one would like to use forces calculated from ab initio electronic structure calculations. Of course, the calculation of ab initio forces is much more expensive, but with the help of currently available massively parallel supercomputers there are many problems that can be tackled this way. We have written an implementation of PRMD that obtains forces on the fly at each simulation step from ab initio packages. This implementation was done by utilizing a glue code, written in Perl, that ties together the PRMD code with an electronic structure code (see Fig. 2). At each step of an MD simulation, the PRMD code passes the current positions of the simulated system to the Perl script, which processes them and sends them to an electronic structure code. After the electronic structure code calculates the current forces, they are passed via the same script back to the PRMD code. Our implementation of the ab initio PRMD method contains two levels of parallelism: first, it parallelizes the molecular dynamics simulation in the time domain by the use of multiple replicas; second, the energies and forces at every step are calculated by the use of parallel electronic structure codes. Ab initio PRMD has a very favorable scaling with respect to the number of processors and, depending on the type of the system, hundreds (and even thousands) of processors can be used to perform the simulations.

The ab initio PRMD was implemented on the Ranger supercomputer at the Texas Advanced Computer Center and applied to the study of diffusion dynamics of helium in hexagonal ice. The Vienna Ab-initio Package Simulation (VASP) program [2, 3] was

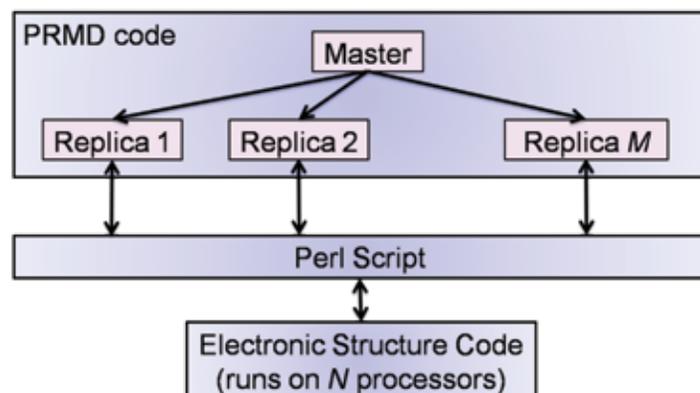


Fig. 2. *Ab initio* PRMD implementation.

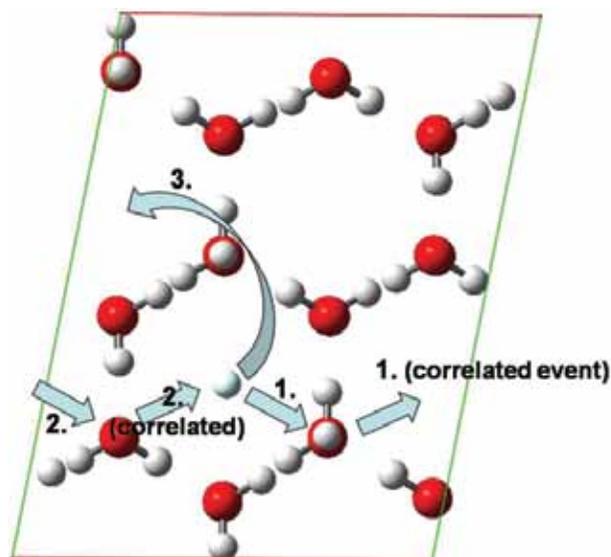


Fig. 3. Helium in hexagonal ice. Blue arrows show observed transitions.

used to obtain forces at every step of the MD simulation. One hundred replicas were used to parallelize the MD simulation in the time domain, with each electronic structure calculation parallelized on 16 processors (16 processors equal one node on Ranger),

leading to the total use of 101 nodes (one for each replica and a master node) corresponding to the use of 1616 processors total. The number of replicas was reduced to 20 in the later stages of the simulation. The preliminary results of the MD simulation are shown in Fig. 3. The simulation was done at 263K, with a 0.5 fs time step. One-picosecond blocks were used for dephasing and decorrelating. The total MD time needed to observe the three transitions depicted in Fig. 3 was 340 ps. With each time step lasting approximately 140 s of wall time, it would take 3.02 yr to reach this time with a standard MD simulation—however, it took only 42 days with our implementation of the *ab initio* PRMD methodology.

In conclusion, we have implemented the *ab initio* PRMD method, which is capable of simulating infrequent event processes. We have tested this implementation by studying diffusion of helium in hexagonal ice and shown that the *ab initio* PRMD is capable of reaching time scales several order of magnitudes longer than standard *ab initio* MD methods while maintaining full atomistic description of the simulated system.

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- [2] G. Kresse, J. Furthmüller, *Comput. Mater. Sci.* **6**, 15 (1996).
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