Extended multicanonical method combined with thermodynamically optimized potential

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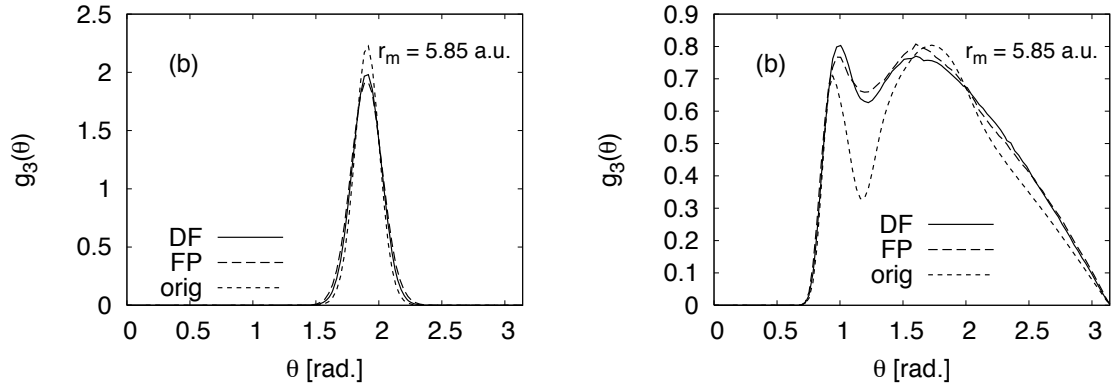
Both the development of the computers and the algorithms are making the first-principles approaches applicable to very large systems. This increase of the number of the atoms in a simulated system will inevitably make both the relaxation time and number of the relevant atomic configurations longer and larger, respectively. Therefore, we also have to pay attention to these two issues to keep calculations well proportioned.

For the increase of the number of relevant atomic configurations, the multicanonical method can be a powerful state survey algorithm that reduces relaxation time of a simulated system and determines its thermodynamic properties in a wide range of temperature.

Therefore, Yoshimoto has studied its combination with first-principles simulations of real materials. The study focused crystal↔liquid transition which is a basic procedure for material synthesis. The talk will present his recent results: a direct simulation of the crystal↔liquid transition by a kind of two-component multicanonical ensemble, a *multi-order multi-thermal ensemble*, with an order parameter defined with structure factors that characterize the transition, and optimization of a model interatomic potential in terms of the ensemble from an accurate one called *thermodynamic downfolding* of a potential. These provide a principle to project a first-principles approach on a model-based approach conserving thermodynamic properties of multiple phases to a maximum extent.

The obtained transition temperature, enthalpy of fusion, and volume change of the transition agreed well with the previous first-principles and the experimental results. It also produced good agreement of the pair-correlation function and the bond-angle distribution for both phases with the first-principles results.



The bond-angle distribution for crystal(left) and liquid(right) phases. DF, FP, and orig mean the results by the downfolded model potential, by first-principles calc., and by origially parametrized model potential, respectively. Each two atoms are considered to be bonded if the distance between them is less than r_m.

[1] Y. Yoshimoto, J. Chem. Phys. 125, 184103 (2006)