TOWARD PREDICTIVE COMPUTATIONAL DESIGN OF PRECISION MOLECULAR OPTOELECTRONICS

Research Challenge
Chemical technology can fabricate solid-state materials made of organic components with precise dimensions for optoelectronic device and catalytic applications. An even greater advance occurs when this synthetic capability is coupled with computational machinery that can predict their properties and functions quantitatively. *Ab initio* many-electron theory provides the necessary foundation, but its legacy algorithms based on matrix algebra are poorly scalable for larger molecules or larger computers. This project introduces completely new and scalable stochastic algorithms.

Methods & Codes
The usual sum-of-products matrix expressions of second-order MBGF (GF2) theory and its complete-basis-set (CBS) correction by explicitly correlated (F12) ansätze are mathematically transformed into high-dimensional integrals, which are then evaluated by a highly scalable Metropolis Monte Carlo (MC) method. The resulting stochastic methods—MC-GF2 and MC-GF2-F12—can compute energy differences directly without a sign problem in a scalable manner with respect to both compute size and system size.

Results & Impact
The developed MC-GF2-F12 method efficiently executes on both XE and XK nodes. It enables an exact (CBS-limit) GF2 calculation of electron-detachment energies for a wide range of large conjugated organic molecules. The largest calculation for C$_{70}$ with 1,610 basis functions was run on 128 GPUs for the GF2 portion and on 896 CPUs for the F12 part. The implemented two-level parallelism enhances the performance of the redundant-walker algorithm on GPUs beyond the degree that is possible by merely running it on many CPUs.

Why Blue Waters
The stability and ease of use of Blue Waters as well as the balanced deployment of CPUs and GPUs are all essential for rapid coding/profiling of new scalable algorithms from scratch and their capacity testing.