

***Ab Initio* Monte Carlo Simulations of Fluid Phase Equilibria at Extreme Conditions**

L.D. Gelb^{C, S}

*Department of Chemistry and Center for Materials Innovation, Washington University in St. Louis, St. Louis,
MO, U.S.A.
gelb@wustl.edu*

We present a simulation framework in which a combination of modern Monte Carlo sampling methods are used in concert with standard electronic structure codes (CPMD and NWChem) to perform liquid-vapor and liquid-liquid coexistence calculations from first principles. The use of an approximate pre-sampling potential to generate large moves with a high probability of acceptance is critical to the method, and our implementation includes on-the-fly refinement of the approximate sampling potential for improved performance. Issues of efficiency, correctness and load-balancing arising from the use of iterative density functional theory potentials are addressed both directly and through analysis of model systems. The method is applied to phase coexistence and (P,V,T) data in simple metals including lithium and sodium, as well as to other systems. The prospect for extension of this technique to more complex systems and solid-liquid phase coexistence is also discussed.