First principles electronic and ionic transport properties across temperature regimes

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In recent years, the Fusion Energy Science Advisory Committee has called on researchers to better understand the state, transport, and dynamic properties of warm dense matter, [1] otherwise known as WDM. More specifically, we have seen a growing interest in the evolution of high energy density (HED) matter as it transits the WDM regime and the hot dense matter (HDM) regime into a burning plasma. Experimentalists are now exploring this phenomenon in a variety of promising ways. For example, laser-based inertial confinement fusion (ICF) experiments have recently used radiation pressure to compress diamond to incredibly high pressures; other experiments are being designed to explicitly test our understanding of how matter mixes in the WDM regime. Other examples come from LCLS, where x-ray scattering was used to study matter in extreme conditions such as observing continuum lowering.

As these experiments push matter into exotic states, where many competing effects are comparable and new theoretical challenges emerge. The regimes in question are hot, but not hot enough to qualify as plasma; they are too dense to be treated as ionized gas, but too hot to be treated as condensed matter. The WDM regime lies in an awkward intermediate region where all the physics is important and competes to determine the physical behavior of the matter; the quantum nature of the electrons remains particularly difficult to model accurately. Because of these challenges, theories interpolate through the WDM regime, connecting the known asymptotic limits at high and low temperature. Commonly used analytic theories have been shown to be dramatically wrong, for example Lee-More and Spitzer thermal conductivity models have significant deviations compared to ab initio results. Such differences in the thermal conductivities have been shown to significantly impact simulation results.

Currently, no single theoretical method (neither orbital based nor the orbital free-Density Functional Theory) can be used to produce consistent, first principles results from ambient conditions through the WDM regime to the HDM regime. Without a better understanding of transport, mixing, and accurate EOS in the WDM and HDM regimes, many aspects of current HED experiments and simulations will remain limited. In fact, this concern was raised in a recent report from the Office of Science and National Nuclear Security Administration: “we urgently need an advanced, finite-temperature many-particle theory that yields the limiting cases of the zero-temperature (condensed matter) and high-temperature (fully to partially ionized plasmas) limits. Such an approach can be combined with molecular-dynamics simulations or integral equation methods to describe disorder and correlations in the WDM state properly. Furthermore a major speedup in these calculations would add significantly to our ability to calculate accurate WDM optical, electrical and thermal conductivities, as well as improved thermodynamic properties." [2]

We propose to merge the best qualities of the orbital-based and orbital-free DFT approaches into a single, unified simulation technique that can consistently handle the full scope of WDM to HDM regimes. At the core, this is a conceptually new DFT approach that avoids previous computational shortcomings in the high temperature limit and therefore can bridge temperature scales. Quantum molecular dynamics (QMD) in general has the advantage that all static, dynamical, and conductive properties arise directly from the density of the electrons and the ions. Such a formulation treats in a consistent manner a system of any composition; thus, the dynamics of mixtures arise in a natural way without any need for ad hoc mixing rules.

Extending theoretical capabilities to produce a consistent, first principles method will have three distinct advantages:

- **Increased range across temperature regimes**. A single, unified method will allow us to expand our simulation range across temperature regimes from ambient through WDM all the way up to HDM.
- **Improved physical accuracy in high temperature regimes**. The proposed method will retain the quantum nature of localized, core electronic states, thus retaining their impact in the WDM and HDM regimes. This will allow for a more accurate description of the electrons in a range of parameter space where such physics has largely been omitted, including continuum lowering and ionization features. This work will allow for a single method to produce essentially all the microscopic properties over a large range of phase space in a consistent manner. For example, retaining the quantum nature of the electrons will enable calculations of conductivities from first principles for the WDM and HDM regimes. Such a tool will naturally apply to the study of mixtures.
- **Improved computational efficiency and impact**. This new method will reduce the computational cost of DFT in the high temperature regimes, and allow QMD to be applied in the WDM and HDM regimes. Thus providing consistent transport properties for the first time across these temperature regimes. It will provide first principles, consistent EOS and conductivities to hydrodynamics simulations and kinetic simulations relevant to the DOE.

The proposed efficient QMD method will allow us to study mixtures of interest to the Fusion Energy Science Program. We intend to develop a theory of WDM mixtures (multiple species of ions) with extensive studies of the fuel and ablator systems, including EOS, mass transport properties, and conductivities. The accurate tabulation of such properties will be at the heart of advancing Inertial Fusion Energy (IFE) simulations through high performance computing. Progress in QMD will be necessary to understanding of x-ray scattering, including Thomson scattering. Ultimately all systems used to search for and achieve fusion will be composed of mixtures. Therefore, it is essential that as experiments progress, we develop a truly predictive capability that can span all required temperature regimes and produce consistent results. This will allow for more accurate simulations of HED matter, paving the way for fusion energy.

References

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