White Paper for *Frontiers of Plasma Science Panel*

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Primary (P) and secondary (S) areas this white paper addresses

•Plasma Atomic physics and the interface with chemistry and biology	S
•Turbulence and transport	S
•Interactions of plasmas and waves	
•Plasma self-organization	
•Statistical mechanics of plasmas	P

Indicate type of presentation desired at Town Hall Meeting

Oral	X
Poster	
Either Oral or Poster	
Will not attend	

Title:	First-principles Simulations of High Energy Density Matter
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1 Describe the research frontier and importance of the scientific challenge.

A rigorous and consistent theoretical description of materials residing in the high energy density physics (HEDP) and warm dense matter (WDM) regimes has been identified [1, 2] as a central goal to the development of key plasma technologies, such as inertial and magnetically confined fusion, shock physics, high energy astrophysics, and stockpile stewardship. Some of the most important pieces of information theoretical calculations can provide include the equation of state (EOS), which relates energy, pressure, density, and temperature to describe states of matter, and transport and optical properties, such as viscosity, diffusivity, conductivity and opacity. The EOS and transport properties provide fundamental information from which many other thermodynamic properties can be derived and, subsequently, used to build theoretical models and design HEDP experiments. For example, the design of a fusion experiment relies heavily on EOS models and predicted transport and optical properties to optimize the design parameters such that it can produce the pressures and temperatures required for ignition. In addition, the EOS and transport and optical properties constrain key input parameters in widely-used hydrodynamic simulations of plasmas, which also further reduce experimental uncertainties and hydrodynamic instabilities in the designs of experiments. Therefore, investing resources into the theory and computation of highly accurate equations of state and transport and optical properties is crucial for achieving the goals of the Plasma Science Frontiers program.

The main challenge to theorists comes from the fact that a comprehensive first-principles framework that can accurately and reliably compute equations of state across the entire WDM regime does not yet exist. While there are several complementary methods in development, more work needs to be done applying current methods to computing transport and optical properties and developing the same capability in others. In the HEDP regime, materials are generally considered to exist as fully ionized plasmas, which are well-described by a variety of analytic physics models.

In the relatively colder WDM regime, materials are only partially ionized, and electrons are still strongly correlated. Hence, in the WDM regime, quantum effects, such as quantum degeneracy, ionization, and dissociation are all important in determining the equation of state [3]. It is because of these effects that a first-principles treatment of the WDM regime is challenging and that an efficient computational method is still needed to reliably compute equations of state and transport and optical properties.

2 Describe the approach to advancing the frontier and indicate if new research tools or capabilities are required.

There are several first-principles-based approaches under development for predicting equations of state and transport and optical properties for plasmas. A concise discussion of the current state of the most prominent methods was recently published set of lectures [4]. Here, we briefly summarize the strengths, weaknesses, and accomplishments of the major approaches.

Currently, the most widely-used and accessible first-principles method for studying WDM is Kohn-Sham density functional theory (KS-DFT), which maps the many-body electron problem onto an efficiently solvable single-particle problem. For WDM studies, KS-DFT molecular dynamics treats the electronic temperature in a perturbative approach by allowing for thermal smearing of the band occupation, and treats ion-motion as purely classical. The main drawback of KS-DFT-MD is that it becomes computationally infeasible as temperature increases and the number of occupied bands becomes too unwieldy to process. In general, KS-DFT-MD is capable of producing accurate EOSs and transport and optical properties for temperatures less than 100 eV.

There are at least three outstanding methods at the frontier of first-principles WDM prediction of EOS and transport and optical properties, which aim to go beyond standard thermal KS-DFT-MD:

(1) The Path integral Monte Carlo (PIMC) method [5], which is the method developed by our group, is a high-accuracy, many-body alternative to the DFT-based methods. While DFT-based methods treat finite temperatures in an approximate manner, PIMC naturally and correctly incorporates finite temperatures of electrons and ions by working within the full many-body thermal density matrix formalism and mapping it onto a stochastic process. The only uncontrolled approximation is that of the nodal surface to deal with the fermion sign problem. While PIMC has most often been applied to hydrogen and helium with free-particle nodes, recent developments have shown that, in combination with DFT-MD, PIMC has great potential to construct a coherent EOS for heavier elements throughout the WDM and HEDP plasma regimes. [6, 7, 8]. We are also making promising progress on development of localized nodal surfaces for heavier elements. Development is still needed to compute transport and optical properties from PIMC.

(2) Orbital-free DFT approaches dramatically improve the efficiency of DFT-MD's temperature scaling by approximating the noninteracting free energy by a functional instead of computing it from single particle orbitals [9]. In general, this speed-up has resulted in an significant trade-off in accuracy, but recent OF-DFT developments have shown the method is potentially capable of being competitive with KS-DFT accuracy [10, 11]. OF-DFT is still limited to simulations of a few hundred particles and relies on a pseudopotential and exchange-correlation approximations but is also capable of computing transport and optical properties [12].

(3) In an effort to make even greater gains in efficiency, DFT-based average-atom models make further approximations in which the properties of a single atom are solved for within the plasma [13]. Such models have been shown to predict the electronic structure of the atom well, and recent developments have begun a more consistent treatment of ionic structure that allow it

to achieve accuracy on par with OF-DFT [14] and provide transport and optical properties [15].

Other methods that focus on accurate computation of dynamical and transport properties and response functions include time-dependent DFT, many-body perturbation theory, dynamical structure factor calculations, and quantum electro dynamics.

3 Describe the impact of this research on plasma science and related disciplines and any potential for societal benefit.

The continued development of first-principles models for the computation of EOS, transport, and optical properties in the areas of WDM and HEDP will have a large impact in the areas of fusion energy, planetary science, stellar modeling, astrophysics, and stockpile stewardship. One of the most important impacts such research could have is in the updating of EOS databases that are based on out-dated, semi-analytic models. Unless direct measurements are available, all such semi-analytical models have limited predictive capabilities. It is not unusual for the density to be off by 10-50% in the regime of warm dense matter where plasma models do no longer work because the coupling between particles is strong. The SESAME [16] and the QEOS [17] models were all developed more than two decades ago when first-principles simulations could only access a limited range of pressures and temperatures and CPU time was a scarce resource. With current computing technology and resources available, it is time to develop a mature, first-principles simulation tools for WDM and HEP EOS computation for many different materials over a large range of T and P conditions. The ability to overhaul the semi-analytic EOS models and develop a first-principles EOS database will be valuable to the modeling, design of new experiments, and the stewardship of plasma science.

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