Warm Dense Matter: A hot topic at the frontiers of Science

Ab Initio Simulation of Warm Dense Matter

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In Short

- Warm Dense Matter (WDM) is an extreme state that occurs, for example, in astrophysical objects and on the pathway towards inertial confinement fusion
- State-of-the art WDM experiments at large research facilities like the European XFEL require rigorous theoretical support
- Density functional theory (DFT) constitutes the defacto work-horse of WDM, but external input from other theories is required
- Quantum Monte Carlo (QMC) methods provide the best description of Warm-dense matter and are essential for the development of DFT simulations of WDM

Warm dense matter (WDM) is an extreme state of matter characterized by high densities and temperatures and has emerged as one of the most challenging frontiers of plasma physics and material science [1,2]. These conditions occur in many astrophysical objects such as in the interiors of giant planets, in brown dwarfs, and in neutron star crusts, see Fig. 1 for an overview. Moreover, they occur in inertial confinement fusion capsules on their pathway towards ignition [3] and are potentially relevant for understanding radiation damage in both fission and fusion reactor walls. Furthermore, they are relevant for the novel field of hot-electron chemistry where the latter are used to accelerate chemical reactions.

These applications have sparked a surge of activities in this field, and WDM is now routinely realized in large research centers around the globe such as the European XFEL in Schleswig-Holstein/Hamburg that went into operation only recently. This has lead to a multitude of experimental breakthroughs, such as the discovery of diamond formation in compressed carbon [4], which might have profound implications for the structure and evolution of icy giant planets like Neptune and Uranus.

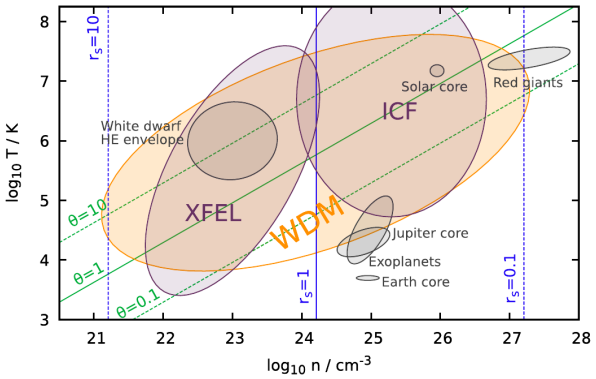

Figure 1: *Schematic overview of WDM applications in the density–temperature plane. Note that XFEL denotes conditions that can be realized with free electron lasers like the new European XFEL, and ICF corresponds to inertial confinement fusion [3]. Moreover, r_s and θ denote the density parameter and degeneracy temperature, see Refs. [1,2]. Taken from [2].*

At the same time, the interpretation of these experiments requires the availability of accurate theoretical models to obtain important plasma parameters like the electronic temperature and density that cannot be directly measured. Yet, the theoretical modelling of WDM constitutes a formidable challenge due to the complicated interplay of 1) Coulomb repulsion, 2) thermal excitations, and 3) quantum degeneracy effects (like Pauli blocking of the electrons). For these reasons, thermal density functional theory has emerged as the most widely used tool for WDM simulations as it combines an—at least in theory—*ab initio* treatment of the electrons with a computational cost that is manageable on large computer clusters such as HLRN.

In this context, the central problem of DFT is the choice of the so-called *exchange–correlation* (XC) functional, which cannot be obtained within DFT itself and has to be supplied as an external input. At ambient conditions, where the electrons are in the ground state, sophisticated XC functionals have been constructed on the basis of ground-state QMC simulations of the uniform electron gas (UEG) [5]. This, in turn, has enabled the unprecedented success of DFT regarding the description of real materials in physics, quantum chemistry, and beyond.

Here, we aim to extend this success to the field of WDM research. First and foremost, we mention that the application of the aforementioned ground-state functionals at high temperature leads to substantial errors [6], so that the development of novel, explicitly thermal XC-functionals is paramount. As a first step, we have recently obtained a full, highly

accurate parametrization of the XC free energy of the UEG [7] covering the entire relevant density–temperature range. This was achieved on the basis of novel QMC simulation techniques [8,9,10,11] developed by our group.

In the present project, we will significantly extend our previous considerations by utilizing the unique capabilities of the groups from Kiel and Dresden regarding both thermal DFT and thermal QMC methods [2]. First and foremost, we will use QMC data for the XC-properties of the warm dense UEG [12] to construct advanced XC-functionals that are specifically designed for WDM applications [13]. In this regard, an additional use of our QMC methods for the further development of thermal DFT simulations is the availability of highly accurate benchmark data. This will allow for the first time to rigorously assess the accuracy of different XC-functionals in the WDM regime.

An additional goal of this project is the large-scale thermal DFT simulation of realistic WDM applications. This includes the investigation of plasmonic excitation that are of key relevance for the interpretation of X-ray Thomson scattering experiments, or real time-dependent DFT calculations for the stopping power.

Moreover, QMC simulations of electrons in the WDM regime are very interesting in their own right. For example, we have recently used path-integral Monte-Carlo (PIMC) calculations to obtain the first *ab initio* data for the *dynamic* structure factor $S(q,\omega)$ going from WDM to the strongly coupled electron liquid regime [14]. Furthermore, our QMC methods allow to study nonlinear effects in WDM [15], which has potentially important implications for the interpretation of state-of-the-art experiments. Finally, we have recently presented the first *ab initio* QMC results for warm dense hydrogen without the usual fixed node approximation [16].

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More Information

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