First-principles studies on the equation of state, thermal conductivity, and opacity of deuterium–tritium (DT) and polystyrene (CH) for inertial confinement fusion applications

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Abstract. Using first-principles (FP) methods, we have performed ab initio compute for the equation of state (EOS), thermal conductivity, and opacity of deuterium–tritium (DT) in a wide range of densities and temperatures for inertial confinement fusion (ICF) applications. These systematic investigations have recently been expanded to accurately compute the plasma properties of CH ablators under extreme conditions. In particular, the first-principles EOS and thermal-conductivity tables of CH are self-consistently built from such FP calculations, which are benchmarked by experimental measurements. When compared with the traditional models used for these plasma properties in hydrocodes, significant differences have been identified in the warm dense plasma regime. When these FP-calculated properties of DT and CH were used in our hydrodynamic simulations of ICF implosions, we found that the target performance in terms of neutron yield and energy gain can vary by a factor of 2 to 3, relative to traditional model simulations.

1. Introduction
Accurate plasma properties of deuterium–tritium (DT) fuel and ablators under extreme conditions are crucial for simulating and understanding inertial confinement fusion (ICF) implosions. Knowledge of these physical properties, such as equation of state (EOS), thermal conductivities, viscosity, and opacity, is required beforehand to simulate ICF. Historically, these quantities have been estimated with approximated physics models such as the SESAME EOS model [1] the Lee–More [2] or Purgatorio [3] models for thermal conductivity $\kappa$, and the Astrophysics Opacity Table (AOT) [4] for radiation transport. Since the ignition designs generally have small margins, it is legitimate to ask whether or not the high-energy-density plasma properties given by these physics models are accurate enough for reliable ICF target designs. To that end, we have employed first-principles (FP) methods to systematically investigate the EOS, $\kappa$, and opacity of ICF-relevant materials (DT and CH ablator) in a wide range of plasma conditions through state-of-the-art ab initio calculations. Through these
systematic studies, we have established the first-principles equation-of-state (FPEOS) tables [5–8] and thermal conductivities [9,10] (κ_{QMD}) for both DT and CH, as well as the first-principles opacity table (FPOT) [11] for DT. Implementing these FP-based plasma properties [FPEOS, κ_{QMD}, and FPOT] into our hydrocodes, we have examined their effects on ICF simulations. It was found that neutron yield and target gains from hydrodynamic simulations using these FP-based plasma properties can differ by a factor of 2 to 3 with respect to the traditional model predictions [12].

2. The first-principles methods

These FP methods include the quantum-molecular dynamics (QMD) and the path-integral Monte Carlo (PIMC) simulations. Both methods treat the electrons quantum-mechanically, while the ions are described by classical Newtonian mechanics (in QMD). In the PIMC method [13], the density matrix of a many-body system at high temperatures is built from the plane waves that describe the free particles. The density matrix at low temperatures can be obtained by performing the path-integral along the temperature path, using the known high-\(T\) density matrix. This method can handle plasmas with temperatures down to ~0.1 \(T_F\) (\(T_F\) is the Fermi temperature); however, for lower-\(T\) plasmas, the Fermi-sign problem renders the PIMC method impractical.

For complex plasmas (beyond DT) and transport properties of DT, we switched to the QMD method, which is based on the density functional theory (DFT) [14]. The QMD method combines the Kohn–Sham orbital-based molecular dynamics (KSMD) and the orbital-free molecular dynamics (OFMD) [15]. Both methods apply the DFT to find the thermodynamic-equilibrium electron density for each evolving ion configuration in the plasma. Subsequently, the motion of ions is then driven by the electronic force (derived from the obtained electron density) and the ion–ion interaction force, through the classical Newton equation. Once the plasma evolution is simulated, the various plasma properties can be extracted self-consistently. Combining both the KSMD and the OFMD methods, we can sample a wide range of temperature and density conditions relevant to both shell and coronal plasmas in ICF implosions.

3. FP-based plasma properties of DT and CH

In this section, we compare the plasma properties extracted from our first-principles calculations with both experiments and traditional model predictions. To be specific, we have conducted both PIMC and QMD simulations of D\(_2\) plasmas of \(\rho = 0.001\) to 1596 g/cm\(^3\) and \(T = 1000\) K to 64,000,000 K, and CH plasmas of \(\rho = 0.1\) to 100 g/cm\(^3\) and \(T = 1000\) K to 4,000,000 K with QMD calculations. These \(\rho,T\) conditions fully cover the ICF plasma conditions. By doing mass/density scaling to the D\(_2\)/CH results, we can obtain the plasma properties of DT and deuterated plastic (CD). It is noted that the same QMD method can also be directly used for DT/CD property calculations, which should be equivalent to what obtained by mass/density scaling from the D\(_2\)/CH results.

Next, in figure 1, we compare the FP-predicted Hugoniots with experimental measurements [16–21] and the SESAME and Kerley03 models [1,22], respectively, for (a) D\(_2\) and (b) CH.

![Figure 1. Comparison of FPEOS-predicted Hugoniot with both experiments and models.](image-url)
Figure 1 shows that the FP calculations of shock Hugoniot of both D2 and CH agree well with experiments, while the traditional EOS models deviate from the FP predictions in certain pressure ranges. The off-Hugoniot EOS has been compared with the EOS models in previous publications [5–8], which indicated significant differences in warm dense plasma conditions where strong-coupling and electron degeneracy effects are important.

Figure 2 compares the resulting (a) opacity and (b) $\kappa_{\text{QMD}}$ of D$_2$ and (c) the $\kappa_{\text{QMD}}$ of CH with traditional models used in our hydrocodes. We see from figure 2 that the thermal conductivities of warm dense D$_2$ and CH plasmas are higher than the Lee–More predictions by a factor of 2 to 10; also, the total Rosseland opacities of D$_2$ are higher than the cold-opacity patched AOT by a factor of 10 to 100 at plasma temperatures below 20eV.

Figure 2. Comparisons of FP-predicted $\kappa$ and opacity with traditional model predictions.

4. Effects of FP-based plasma properties on ICF simulations

Incorporating these FP-based properties [FPEOS, $\kappa_{\text{QMD}}$, and FPOT] of D$_2$ and CH into our hydrocodes, we can examine their effects on direct-drive ICF designs. One example is shown in figure 3, where a direct-drive ICF target design (a) is simulated with our 1-D hydrocode LILAC. The simulation results are compared figure 3(b), in which the dashed line represents the traditional simulations in contrast to the solid lines using the FP-based plasma properties of DT, while the high-density carbon ablator is still simulated with traditional models. The final target gain varies from $G = 28$ in the traditional models to the more-reliable value of $G = 11$ using the accurate FP-based plasma properties of DT. The effects of CH FPEOS and $\kappa_{\text{QMD}}$ of CH further decrease the target performance [8,10] in CH-ablator based ICF implosions because of the reduction in the implosion velocity. To recover high gains, the FP-based plasma properties should be used to retune the laser pulse shape [12].

Figure 3. Comparisons of hydro simulations of a direct-drive ICF target design, using the FP-predicted properties of DT versus traditional models.
5. Conclusion
Using first-principles methods, we have systematically calculated plasma properties for a wide range of densities and temperatures of DT and CH for ICF applications. The calculated properties have shown significant differences when compared to traditional model predictions. Hydro simulations of ICF implosions with these FP-based plasma properties have shown a gain that is a factor of \(2\times\) to \(3\times\) smaller with respect to traditional model simulations. These studies indicated that FP-based plasma properties of DT and ablators should be used for reliable ICF target designs.

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