Simulating Warm Dense Plasmas with a Hybrid Molecular Dynamics – Quantum Hydrodynamics Approach

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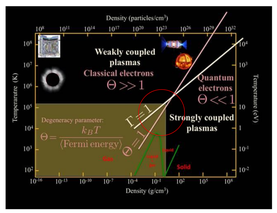
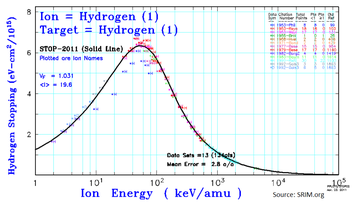
Abstract

Modeling the slowing-down of charged projectiles in warm dense matter (WDM) is of great interest to the design of ion-beam and intense laser experiments, e.g., in describing fast ion-heating in ICF. This process is complicated by the electrons being moderately Coulomb-coupled and partially quantum-degenerate, while also dynamically screening the projectile. In this regime, ab initio Molecular Dynamics (MD) simulations are typically employed, and solve for the self-consistent quantum ground-state electron density for the ion configuration at each timestep using Density Functional Theory (DFT). Here, we describe a Quantum Hydrodynamics (QHD) approach, which treats the electrons as a fluid with forces derived from DFT, ensuring accurate equilibrium equation of state properties. We derive the QHD equations from first principles, connect them to the machinery of DFT, and describe the predicted linear response. We developed a parallelized simulation code that combines MD ions and QHD electrons, and simulated a stopping power experiment conducted at the Jupiter Laser Facility. By comparing with a quantum statistical potential MD code, we show favorable results for the QHD-MD approach. We discuss possible extensions into the FLASH code framework.

Motivation

WDM, characterized by moderate Coulomb coupling $\Gamma \sim 1$ and partial quantum degeneracy $\theta \sim 1$, is a "barrier in parameter space" where well-defined models break-down. For example, free molecules in condensed matter experience bond-breaking/ionization with increased pressure, while plasmas have Debye screening approaching inter-particle spacing with lower temperatures. A process of great interest is energy deposition of charged projectiles, as in bootstrap heating of fuel during thermonuclear burn. Projectiles near the electron thermal velocity have a maximum in stopping, the Bragg peak, which requires careful bound state calculations. We want to model early stages of ICF when the capsule is in the WDM regime, so the method must effectively treat quantum as well as dynamical electrons.





The purpose of this work is to develop a physical model and direct numerical simulation capability that is applicable over a broad range of parameter space, specifically motivated by the problem of charged particle stopping in warm dense matter. Ions must be dynamical, and may be strongly Coulomb-coupled, thus will be treated as classical point particles with molecular dynamics. Electrons must be quantum mechanical, and to ionize/capture and make no assumptions about bound/core electronic states, will incorporate techniques of orbital-free density functional theory. Electrons must also be dynamical yet computationally tractable, and to be able to nonlinearly screen the charged projectiles, will be treated hydrodynamically. This work combines a molecular dynamic description of ions with a quantum hydrodynamic model for electrons.

Quantum Hydrodynamics Theory

I derived a many-body QHD model from first principles in the spirit of Madelung. The exact, time-dependent, 3-d fluid equations enforce wave function anti-symmetry, and to obtain a simple closed form, I assume a Slater determinant wave function, with plane-wave orbitals in the uniformly-dense limit. This model seeks to clarify the assumptions inherent in the QHD picture and put it on a more rigorous footing.

$$\begin{aligned} \frac{\partial n(\mathbf{r})}{\partial t} + \nabla\cdot\mathbf{j}(\mathbf{r}) &= 0, \\ m\,\frac{\partial \mathbf{j}(\mathbf{r})}{\partial t} + m\,\nabla\cdot\left(\frac{\mathbf{j}(\mathbf{r})\,\mathbf{j}(\mathbf{r})}{n(\mathbf{r})}\right) &= -\nabla\cdot\Big(\mathbf{P}(\mathbf{r}) + \mathbf{Q}(\mathbf{r})\Big) - n(\mathbf{r})\,\nabla\Phi(\mathbf{r}), \end{aligned}$$

$$\begin{aligned} \mathbf{P}(\mathbf{r}) + \mathbf{Q}(\mathbf{r}) &= \frac{\hbar^2}{m}\,\frac{(3\pi^2)^{2/3}}{5}\,n(\mathbf{r})^{5/3}\,\mathbf{I} - \lambda\,\frac{\hbar^2}{4m}\,n(\mathbf{r})\nabla\nabla\ln n(\mathbf{r}), \\ \Phi(\mathbf{r}) &= -e^2\left(\frac{3\,n(\mathbf{r})}{\pi}\right)^{1/3} + e^2\int d\mathbf{r}'\,\frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + V(\mathbf{r}). \end{aligned}$$

A separate approach to QHD is with a phenomenological Bloch model, where electrons are assumed to satisfy Euler equations with a chosen equation of state. Ying was first to use the chemical potential from Density Functional Theory. Since the DFT energy functional is stationary in the ground state, it has the desirable property of relaxing the hydrodynamic system to the quantum ground state solution.

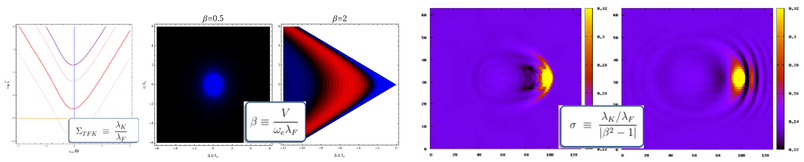
$$\begin{aligned} \frac{\partial n}{\partial t} + \nabla\cdot n\mathbf{v} &= 0, \\ m\,n\left(\frac{\partial}{\partial t} + \mathbf{v}\cdot\nabla\right)\mathbf{v} &= -n\,\nabla\mu \end{aligned}$$

$$\begin{aligned} \mu(\mathbf{r}) &= \frac{\delta T[n(\mathbf{r})]}{\delta n(\mathbf{r})} + \frac{\delta \mathcal{F}_{xc}[n(\mathbf{r})]}{\delta n(\mathbf{r})} + e^2\int d\mathbf{r}'\,\frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + V(\mathbf{r}). \\ \frac{\delta T[n(\mathbf{r})]}{\delta n(\mathbf{r})} &= \eta\,T + \frac{\hbar^2}{m}\left[\left(h(\eta) - n\,\frac{dh(\eta)}{dn}\right)\left|\frac{\nabla n}{n}\right|^2 - 2\,h(\eta)\,\frac{\nabla^2 n}{n}\right], \\ \eta(\mathbf{r}) &= \mathcal{I}_{1/2}^{-1}\left[\frac{\pi^2}{\sqrt{2}}\left(\frac{\hbar^2}{mT}\right)^{3/2} n(\mathbf{r})\right], \quad h(\eta(\mathbf{r})) \equiv \frac{\mathcal{I}_{1/2}(\eta)\,\mathcal{I}'_{-1/2}(\eta)}{12\,\mathcal{I}'^2_{-1/2}(\eta)}. \\ \frac{\delta \mathcal{F}_{xc}[n(\mathbf{r})]}{\delta n(\mathbf{r})} &= -e^2\left(\frac{3n}{\pi}\right)^{1/3} + \frac{\delta \mathcal{F}_c[n(\mathbf{r})]}{\delta n(\mathbf{r})}. \end{aligned}$$

Comparing the two models justifies using Euler equations for electron dynamics, with forces from the external, Hartree mean-field Coulomb, and Dirac exchange potentials, while the Thomas-Fermi chemical potential replaces the pressure, and the Kirzhnits gradient correction is analogous to the Bohm pressure.

Linearized QHD

Linearizing the Bloch QHD equations produces two natural length scales: the Thomas-Fermi screening length and a diffractive Kirzhnits length. I define a diffractive parameter Σ as the ratio of Kirzhnits to Fermi lengths, which determines the character of solutions. $\Sigma < ½$ (non-diffractive) gives exponential screening, while $\Sigma > ½$ (diffractive) has stationary oscillatory solutions related to Friedel oscillations. The induced density around a charged projectile can be solved analytically. The first parameter of interest is β, the ratio of projectile to Fermi velocites. $\beta < 1$ (sub-thermal) gives exponential screening on elliptical surfaces, while $\beta > 1$ (super-thermal) produces a conic wake with oscillatory hyperbolic surfaces. Another parameter of interest is σ, a velocity-dependent diffractive parameter. $\sigma < ½$ (non-diffractive) behaves similarly to TF, while $\sigma > ½$ (diffractive) generates oscillations ahead of the projectile.



Plotted first are contours of Σ in Coupling-Degeneracy space, with purple the critical value ½. Next is TF screening density for sub- and super-thermal β. Lastly is TFK screening for non-diffractive and diffractive σ. It is important to note that diffractive effects are greatly enhanced near the Fermi velocity, or Bragg peak.

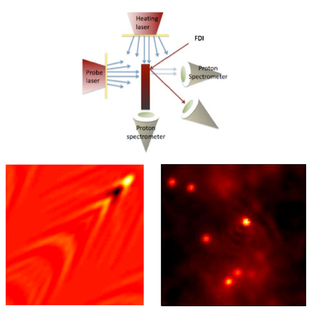
Numerical Methods

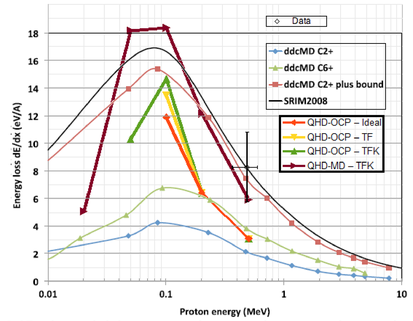
NEREID (N-body Energy-functional hydRo for Electron-Ion Dynamics) is a 3-dimensional hybrid particle/fluid code I developed to simulate QHD problems. It is written in C and parallelized with MPI. Inter-particle forces use P^3M, where the interactions are Ewald-decomposed into short-range P-P solved directly, and long-range P-M using FFTs on a mesh. Molecular Dynamics uses Velocity-Verlet, with forces from P^3M and the electron fluid on the mesh. Hydrodynamics uses a Finite-Volume Method in cartesian coordinates, with fluxes from Quadratic Upstream Interpolation for Convective Dynamics, and time integration with a Predictor-corrector method. The Equation of State for the electron fluid can be Thomas-Fermi potential or pressure, with Kirzhnits gradient-correction, and Dirac exchange.

FLASH is a high-performance multiphysics code capable of handling general compressible flow problems found in plasma and astrophysics environments. Work is under-way to implement a QHD solver into the FLASH framework.

Stopping Power Simulations

An experiment was conducted at the Jupiter Laser Facility to measure proton stopping in WDM carbon. A graphite target was heated to T = 13 eV and irradiated with a proton beam energy E = 500 keV, then a proton spectrometer measured a stopping force dE/dx = 84±20 eV/nm. This experiment was simulated for a range of projectile velocities using several codes. SRIM (Stopping Range of Ions in Matter) is a group of programs simulating binary scattering events with potentials from quantum calculations and experimental data. The classical molecular dynamics code ddcMD uses Quantum Statistical Potentials that smooth the short-range interaction to account for some quantum behavior. NEREID was run in two ways: QHD-OCP simulated the single ion projectile in the free electron fluid (<Z> ~ 2.2), and QHD-MD simulated the projectile and all bare carbon nucleii in the total electron fluid.





Stopping power calculated by QHD-MD is within experimental error bounds and agrees relatively well with SRIM for velocities near and above the Bragg peak, validating this approach for direct numerical simulation of stopping in WDM.

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