# Kinetic theory molecular dynamics and hot dense matter: Theoretical foundations

F. R. Graziani, J. D. Bauer, and M. S. Murillo

Phys. Rev. E **90**, 033104 — Published 3 September 2014

DOI: 10.1103/PhysRevE.90.033104

# Kinetic Theory Molecular Dynamics and Hot Dense Matter: Theoretical Foundations

F. R. Graziani and J. D. Bauer

*Lawrence Livermore National Laboratory*

*Livermore, CA 94551, U.S.A.**

M. S. Murillo

*Los Alamos National Laboratory*

*Los Alamos, NM 87544, U.S.A.*

(Dated: August 19, 2014)

## Abstract

Electrons are weakly coupled in hot, dense matter that is created in high-energy-density (HED) experiments. They are also mildly quantum mechanical and the ions associated with them are classical and may be strongly coupled. In addition, the dynamical evolution of plasmas under these hot, dense matter conditions involve a variety of transport and energy exchange processes. Quantum kinetic theory is an ideal tool for treating the electrons but it is not adequate for treating the ions. Molecular dynamics is perfectly suited to describe the classical, strongly coupled ions but not the electrons. We develop a method that combines a Wigner kinetic treatment of the electrons with classical molecular dynamics for the ions. We refer to this hybrid method as "kinetic theory molecular dynamics," or KTMD. The purpose of this paper is to derive KTMD from first principles and place it on a firm theoretical foundation. The framework that KTMD provides for simulating plasmas in the hot, dense regime is particularly useful since current computational methods are generally limited by their inability to treat the dynamical quantum evolution of the electronic component. Using the $N$-body von Neumann equation for the electron-proton plasma, three variations of KTMD are obtained. Each variant is determined by the physical state of the plasma (e.g. collisional versus collisionless). The first variant of KTMD yields a closed set of equations consisting of a mean-field quantum kinetic equation for the electron one-particle distribution function coupled to a classical Liouville equation for the protons. The latter equation includes both proton-proton Coulombic interactions and an effective electron-proton interaction that involves the convolution of the electron density with the electron-proton Coulomb potential. The mean-field approach is then extended to incorporate equilibrium electron-proton correlations through the Singwi-Tosi-Land-Sjolander (STLS) ansatz. This is the second variant of KTMD. The STLS contribution produces an effective electron-proton interaction that involves the electron-proton structure factor, thereby extending the usual mean-field theory to correlated but near equilibrium systems. Finally, a third variant of KTMD is derived. It includes dynamical electrons and their correlations coupled to a MD description for the ions. A set of coupled equations for the one-particle electron Wigner function and the electron-electron and electron-proton correlation functions are coupled to a classical Liouville equation for the protons. This latter variation has both time and momentum dependent correlations.



PACS numbers:

*Electronic address: graziani1@llnl.gov

## I. INTRODUCTION

The arrival of new high energy density (HED) physics facilities, such as the National Ignition Facility (NIF) [1], and next-generation light sources, such as the Linac Coherent Light Source (LCLS) [2], has enabled experiments on matter in the warm to hot dense regimes. The design and analysis of these experiments have necessarily motivated the development of new computational approaches [3]. Because of the short time scale associated with the laser-matter interaction, the materials are created in non-equilibrium states. While molecular dynamics is a well-established computational method for describing non-equilibrium dynamics of classical systems [4–7], no quantum analog exists today. In this paper, we present a formalism that provides a basis for such a method.

Hot dense matter (HDM) is characterized by temperatures ranging from hundreds of eV to a few hundred keV and densities ranging from tenths to hundreds of gm/cc [8–10]. Hot dense matter conditions occur in stellar cores, Z-pinch, and inertial confinement fusion experiments. These experiments frequently involve non-equilibrium states where multi-ion species undergo thermonuclear burn, collisional, radiative and atomic processes [9, 10]. Examining the plasma coupling parameter $\Gamma$ [11, 12], and the degeneracy parameter $\Theta$

$$\Gamma_{ij} = \frac{Z_i Z_j e^2}{kT a_{ij}} \tag{1}$$

$$\Theta = E_F/kT \tag{2}$$

Where $Z_i$ is the effective ionization of species "i", "e" is the electron charge, k is Boltzmann's constant, T is the temperature of the plasma and $a_{ij}$ is the multi-species ion sphere radius of species "i" and "j".

$$a_{ij} = (a_i + a_j)/2 \tag{3}$$

Low-Z species in multi-ionic HDM tend to be weakly coupled ($\Gamma \ll 1$) while higher Z species can be strongly coupled ($\Gamma \geq 1$). Electron degeneracy is not a concern for the density and temperature conditions we consider. This means that, for densities of 10 gm/cc, temperatures must be higher than 120 eV, and for densities on the order of 100 gm/cc, temperatures must be in excess of 560 eV.

Although degeneracy is not an issue, electrons in HDM do exhibit a quantum characteristic: diffraction. At large distances, Debye screening occurs as a result of classical collective effects. At small distances, the relevant scale that characterizes close encounter Coulomb collisions between electrons and protons and electrons and electrons is the thermal de Broglie wavelength. We will refer to the fact that the electrons in HDM exhibit both classical and quantum behavior as the classical-quantum dichotomy. Additional quantum effects such as atomic transitions and thermonuclear burn can also be present.

HED experiments are challenging because of the extreme nature of the conditions (hundreds of eV to tens of keV in temperature, densities of many gm/cc, and pressures in the megabar to gigabar regime) and the complicated diagnostics and experiments required to obtain high-quality data [2, 13, 14]. Radiation-hydrodynamic codes are used to model the experiments [13, 14] and are an essential tool in the design process. The codes themselves rely on theoretical models that have rarely been experimentally validated in HDM regimes. Recently, researchers have turned to Molecular Dynamics (MD) to validate by simulation, a variety of radiation-hydrodynamic code models [3]. MD has turned out to be a useful and very powerful approach to validating models in radiation-hydrodynamic codes. The most recent examples include electron-ion coupling [15–20], stopping power [21], screening of thermonuclear burn [22], and diffusivity [23].

MD methods rely on classical equations of motion to describe the evolution of a many-body system. Given a set of inter-particle potentials, particle positions and momenta evolve in time according to Newton's equations of motion. When applied to HDM, MD maps the quantum many-body system onto a classical many-body system by using an effective force between particles. Depending on the application, MD can employ Born-Oppenheimer methods [24, 25] (e.g., ionic diffusivity) or it may include dynamic electrons explicitly [3, 15–19, 26, 27] (e.g., electron-ion coupling). The emphasis of this paper is on the latter approach where dynamical electrons are important. Currently, all classical MD methods with explicit electrons use quantum statistical potentials (QSP) [28–38] to capture the classical-quantum dichotomy and to model electron and ion dynamics and their interactions. The wave packet MD methods (WPMD) are the exception [39–43]. However, they suffer from wave packet spreading and are less popular than traditional MD methods that use QSP and we do not consider them further. QSP capture the short-distance diffraction and interference properties while at the same time retaining the long-distance classical behavior of Coulomb

potentials. Hence, QSP have been and continue to be the workhorse of MD simulations for HDM. However, the use of these potentials is proven only for MD simulations of equilibrium phenomena like equation-of-state while many of the applications of interest for HDM are non-equilibrium (e.g. electron-ion coupling and ion stopping). Some investigations into HDM non-equilibrium processes have recognized this difficulty and attempted to avoid the problem by running simulations with like charge electrons and ions ([16]). A great deal of progress in the theoretical understanding of hot dense plasmas has been gained by this approach. Unfortunately, a like charge plasma will not tell us about the quantum behavior at small length scales that characterizes HDM. Even though MD with explicit dynamical electrons continues to be applied to HDM, the classical-quantum dichotomy described above continues to be an unsolved problem. The purpose of the work presented in this paper is to propose an alternative approach.

To overcome the classical-quantum dichotomy and the reliance of MD on QSP, Murillo suggested a quantum kinetic theory-molecular dynamics hybrid which we call kinetic theory molecular dynamics (KTMD) [3]. KTMD uses quantum kinetic theory to describe the electron dynamics and MD to describe the classical ion dynamics. Why use quantum kinetic theory to describe the electron dynamics? First, kinetic theory is the simplest way to represent the many electron system with a time-dependent equation in phase space. Second, quantum kinetic theory automatically accounts for quantum diffraction at small length scales. Third, kinetic theory is flexible in that many variants exist for specific scenarios (e.g. quantum Vlasov for collisionless versus quantum Landau for weakly collisional). Fourth, we retain the possibility for fully non-equilibrium distributions. Finally, the KTMD equations are meant to be used in a computer code. There exists a vast resource of methods developed for numerically solving plasma, radiation, charged-particle and neutron transport/kinetic equations. Numerically solving the KTMD equation leverages the existing numerical solution methods from the MD and transport communities.

The purpose of this paper is to derive KTMD from first principles and place it on a firm theoretical foundation. Recently, Michta, Graziani and Surh [44] used reduction operators to derive a classical version of mean-field KTMD from the Liouville equation. The electronic structure in classical KTMD was computed numerically for a given proton configuration and was shown to compare favorably with the results taken from an MD code where the electrons were explicitly treated.

In this paper, we derive KTMD from a fundamental non-relativistic quantum mechanical description of the many-body problem using the N-body von Neumann equation for the electron-proton plasma, where $N = n_e + n_p$, is the sum of the electron and proton numbers and $N$ is the total number of particles in the plasma. The simplification to an electron-proton plasma is merely to make the presentation easier to follow. The derivation presented in this paper can be easily extended to multiple ion species. We show how the N-body von Neumann equation can be mapped to an $n_p$ classical Liouville equation with both classical proton-proton Coulomb interactions and an effective electron-proton interaction and a one-particle quantum electron kinetic equation. The effective electron-proton interaction includes both the mean-field contribution and a contribution resulting from correlations. Ludwig and collaborators [45] have recently considered a similar approach where they treat the ions exactly with MD and an effective ion-ion potential that is dynamically screened by the electrons. A consequence of the derivation of KTMD presented here is that the method does not rely on assumed inter-particle potentials such as QSP. Instead, inter-particle potentials in KTMD are a derived quantity, fully consistent with the dynamics. Assumptions regarding the equilibrium nature of the plasma are not needed within KTMD.

## II. KINETIC THEORY MOLECULAR DYNAMICS AND TRADITIONAL MOLECULAR DYNAMICS METHODS

Molecular dynamics methods can be categorized as either All-Particle-MD (APMD) or Ion-Only-MD (IOMD). APMD treats all particles, electrons and ions, as classical point particles [6] (again, WPMD being the exception). When dynamical electrons need to be simulated, for example, electron-ion coupling, ion stopping in a two component plasma or ionization processes, APMD is used [3, 19, 26, 46, 47]. APMD offer the benefits of robustness and the ease of implementation since they rely only on solving Newton's equations of motion with a fairly simple two-body QSP for all electrons and ions. APMD map the quantum many-body problem onto a classical many body problem using QSP. Unfortunately, the mapping is only proven for equilibrium many body systems [28, 30–32, 38]. .

IOMD methods treat only the ion dynamics with MD. Electronic structure is computed by another method. A commonly used IOMD is Yukawa-MD [48–50] where the ions interact with each through an imposed screened potential. Quantum Molecular Dynamics (QMD) is

a more advanced IOMD that is also commonly used. In QMD the electronic structure is typically computed using density functional theory (DFT) [51, 52]. Both Yukawa-MD and QMD are examples of IOMD where the Born-Oppenheimer approximation is invoked. Viscosity [49, 50] and conductivity [53, 54] are physical processes that are commonly computed using Yukawa-MD and QMD. Recent efforts extending IOMD beyond the Born-Oppenheimer approximation have been made [55, 56]. However, as a general rule the current implementations of IOMD use the Born-Oppenheimer approximation. QMD methods for computing electronic structure are applicable to warm dense matter where the electrons are degenerate and strongly coupled, temperatures are in the eV range and densities vary from 0.1 to 10.0 times solid density. Yukawa-MD methods have been applied to both warm and hot dense matter.

KTMD is an example of an IOMD. Like Yukawa-MD and QMD, the electronic structure is computed using a technique other than MD. However, KTMD is different from Yukawa-MD and QMD. The Born-Oppenheimer approximation is not used. Instead, KTMD computes the time-evolving electronic structure for electrons self-consistently using a quantum kinetic equation. Unlike Yukawa-MD, where a Yukawa potential is imposed, KTMD specifies the nature of the electron-ion interaction through a self-consistent description of the high temperature many-body dynamics. For example, in the mean-field approximation, KTMD computes a self-consistent screening potential through which the ions interact. The self-consistent screening potential depends on the time-evolving local electron density. In addition, quantum diffraction is included.

Unlike QMD, KTMD is designed for high temperatures where the electrons are dynamical, weakly coupled and where kinetic theory is an accurate description of the electronic structure. At high temperatures where the electrons exhibit a classical-quantum dichotomy, KTMD uses a quantum kinetic equation that includes classical collective effects that give rise to screening in addition to quantum diffraction.

## III. THE PLASMA VON NEUMANN EQUATIONS

The von Neumann equation [57, 58] is the quantum analog of the Liouville equation. It describes the evolution of the Wigner transform of the density operator and it represents a full description of the many-particle dynamics of the quantum system. The KTMD method

is based on being able to map the N-particle von Neumann equation to a classical $n_p$ proton Liouville equation with proton-proton and effective proton-electron inter-particle potentials, and a set of quantum kinetic equations for the electrons. In the paper, we demonstrate that such a mapping can be done. We begin with the von Neumann equation for the Wigner transform of the s-particle density operator, which exhibits a BBGKY hierarchy. A set of simple scaling laws along with an expansion in powers of the electron to proton mass ratio, allows us to transform the $n_p$-proton von Neumann equation into a form that looks like the classical $n_p$-proton Liouville equation.

A few assumptions are made in the derivation presented here. First, we are considering only an electron-proton plasma. The formalism can be easily generalized to include any number of ion species. Second, we consider all particles to be fermions. In reality, for a multi-species plasma, the ions may be bosons, in which case, the equal-time anti-commutator relations need to be replaced by equal-time commutator relations. Finally, for simplicity and ease of presentation, we have dropped the spin variables.

### A. Preliminaries

The position ket of every electron and proton in the plasma is defined by

$$\left|r_1 : r_{n_e}; R_1 : R_{n_p}\right\rangle = (1/\sqrt{n_e! n_p!})\hat{\Psi}_e^\dagger(r_1,t)...\hat{\Psi}_{n_e}^\dagger(r_{n_e},t)\hat{\Psi}_p^\dagger(R_1,t)...\hat{\Psi}_{n_p}^\dagger(R_{n_p},t)\left|0_e;0_p\right\rangle \quad (4)$$

where $|0_e;0_p\rangle$ is the vacuum state and $\hat{\Psi}_a^\dagger(r,t)$ and $\hat{\Psi}_a(r,t)$ are the creation and annihilation operators for a charged particle of species type a at a position and time (r,t). The species index "a" is equal to "e" or "p", for electrons and protons, respectively. The annihilation and creation operators obey a set of equal-time canonical anti-commutation relations appropriate for fermions.

$$\{\hat{\Psi}_a(x,t), \hat{\Psi}_b^\dagger(y,t)\} = \delta_{ab}\delta^3(x-y) \quad (5)$$

$$\{\hat{\Psi}_a(x,t), \hat{\Psi}_b(y,t)\} = 0 \quad (6)$$

$$\{\hat{\Psi}_a^\dagger(x,t), \hat{\Psi}_b^\dagger(y,t)\} = 0 \quad (7)$$

The many-body Hamiltonian operator for the plasma interacting via Coulombic forces is described by

$$\begin{aligned}\hat{H}_{plasma} &= \sum_a \int d^3 r_a \hat{\Psi}_a^\dagger(r_a,t)\left[\frac{-\hbar^2}{2m_a}\nabla_{r_a}^2\right]\hat{\Psi}_a(r_a,t) \\ &+ \frac{1}{2}\sum_a\sum_b \int\int d^3 r_a d^3 s_b V_{ab}(r_a - s_b)\hat{\Psi}_a^\dagger(r_a,t)\hat{\Psi}_b^\dagger(s_b,t)\hat{\Psi}_b(s_b,t)\hat{\Psi}_a(r_a,t) \end{aligned} \tag{8}$$

where $a, b$ are the species index and can equal $e$ or $p$. The electron and proton coordinates are defined by $a = e, r_e = r$ and if $a = p, r_p = R$. We have not included either electromagnetic or external fields in the Hamiltonian (8). The dynamics of the many-body plasma are fully described by equation (8) along with the Heisenberg equations of motion.

$$\frac{\partial \hat{\Psi}_a^\dagger(r,t)}{\partial t} = \frac{i}{\hbar}[\hat{H}_{plasma}, \hat{\Psi}_a^\dagger(r,t)] \tag{9}$$

Equations (5) through (9) constitute a complete description of the many-body quantum plasma. They form the basis for the electron and proton von Neumann equations described in the following sections.

### B. Operator Quantum Dynamics

Define the one-particle electron and $n_p$ proton density operators by

$$\hat{\Delta}_e(r + x/2, r - x/2) = \hat{\Psi}_e^\dagger(r + x/2)\hat{\Psi}_e(r - x/2) \tag{10}$$

$$\hat{\Delta}_{n_p} = \hat{\Psi}_p^\dagger(R_1 + \frac{y_1}{2})...\hat{\Psi}_p^\dagger(R_{n_p} + \frac{y_{n_p}}{2})\hat{\Psi}_p(R_{n_p} - \frac{y_{n_p}}{2})...\hat{\Psi}_p(R_1 - \frac{y_1}{2}) \tag{11}$$

Define the one-particle electron and $n_p$ proton phase space density operators as Wigner transforms of the corresponding density operators

$$\hat{F}_e(r,p) = \int \frac{d^3 x}{(2\pi\hbar)^3} e^{\frac{ip\cdot x}{\hbar}} \hat{\Delta}_e(r + x/2, r - x/2) \tag{12}$$

$$\begin{aligned}\hat{F}_{n_p}(R_1,...,R_n,P_1,...,P_n) &= \frac{1}{n_p!}\int \frac{d^3 y_1}{(2\pi\hbar)^3}...\frac{d^3 y_{n_p}}{(2\pi\hbar)^3} e^{\frac{iP_1\cdot y_1}{\hbar}}...e^{\frac{iP_n\cdot y_n}{\hbar}} \\ &\times \hat{\Delta}_{n_p}(R_1 + \frac{y_1}{2} : R_{n_p} + \frac{y_{n_p}}{2}; R_1 - \frac{y_1}{2} : R_{n_p} - \frac{y_{n_p}}{2}) \end{aligned} \tag{13}$$

The one-particle electron and $n_p$ proton phase space density operators are normalized so that the quantum averages, denoted by $\langle ... \rangle$, are defined by

$$\int d^3r d^3p \langle \hat{F}_e(r,p) \rangle = n_e \tag{14}$$

$$\int d^3R_1 ... d^3R_{n_p} d^3P_1 ... d^3P_{n_p} \langle \hat{F}_{n_p}(R_1, ..., R_n, P_1, ..., P_n) \rangle = n_p \tag{15}$$

Differentiating both sides of equation (12) with respect to time and using the Hamiltonian and operator Heisenberg equations given by equations (8) and (9), we obtain the one-electron von Neumann equation,

$$\begin{aligned} \frac{\partial \hat{F}_e(r,p)}{\partial t} + \frac{p}{m_e} \cdot \nabla_{r_e} \hat{F}_e(r,p) \; &= \; \frac{i}{\hbar} \sum_a \int \frac{d^3x}{(2\pi\hbar)^3} \, d^3q \, d^3r_a \, d^3s_a \, e^{i(p-q)\cdot x/\hbar} \\ &\times \; [V_{ea}(r - r_a + \frac{x}{2}) - V_{ea}(r - r_a - \frac{x}{2})] \\ &\times \; \hat{F}_a(r_a, s_a) \hat{F}_e(r,q) \end{aligned} \tag{16}$$

We have ignored exchange terms by making use of the fact that the electrons are non-degenerate. This assumption is made manifest by making the following approximation in the last term of the right hand side of equation (16)

$$\frac{1}{2} \{ \hat{F}_a(r_a, s_a), \hat{F}_e(r,q) \} \approx \hat{F}_a(r_a, s_a) \hat{F}_e(r,q) \tag{17}$$

If we had retained the anti-commutator, equation (16) would be the von Neumann equation valid for degenerate electrons. We stress that equation (16) is still in operator form and hence electron correlations are still present.

Similarly, differentiating both sides of equation (13) with respect to time and using the Hamiltonian and operator Heisenberg equations given by equations (8) and (9), we obtain the $n - p$ proton von Neumann equation.

$$\left(\frac{\partial}{\partial t}+\sum_{i=1}^{n_p}\frac{P_i}{m_p}\cdot\nabla_{R_i}\right)\hat{F}_{n_p} = 2\frac{i}{\hbar}\sum_{i<j}^{n_p}\int\frac{d^3y_i}{(2\pi\hbar)^3}\frac{d^3y_j}{(2\pi\hbar)^3}d^3\tilde{P}_i\,d^3\tilde{P}_j\,e^{i(P_i-\tilde{P}_i)\cdot y_i/\hbar}e^{i(P_j-\tilde{P}_j)\cdot y_j/\hbar}$$
$$\times\ [V_{pp}(R_i-R_j+\frac{y_i-y_j}{2})-V_{pp}(R_i-R_j-\frac{y_i-y_j}{2})]$$
$$\times\ \hat{F}_{n_p}(R_1,...,R_{n_p},P_1,...,\tilde{P}_i,...\tilde{P}_j,...P_{n_p})$$
$$+\ 2\frac{i}{\hbar}\sum_{i}^{n_p}\int\frac{d^3y_i}{(2\pi\hbar)^3}\,d^3p\,d^3\tilde{P}_i\,e^{i(P_i-\tilde{P}_i)\cdot y_i/\hbar}$$
$$\times\ [V_{ep}(r-R_i+\frac{y_i}{2})-V_{ep}(r-R_i-\frac{y_i}{2})]$$
$$\times\ \hat{F}_e(r,p)\hat{F}_{n_p}(R_1,...,R_{n_p},P_1,...,\tilde{P}_i,...P_{n_p}) \qquad (18)$$

A more compact form of equation (18) is

$$\left(\frac{\partial}{\partial t}+\sum_{i=1}^{n_p}\frac{P_i}{m_p}\cdot\nabla_{R_i}\right)\hat{F}_{n_p} = 2\frac{i}{\hbar}\sum_{i<j}^{n_p}V_{pp}(R_i-R_j)\sin\{\frac{\hbar}{2}\{\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i}+\overleftarrow{\nabla}_{R_j}\cdot\overrightarrow{\nabla}_{P_j}\}\}\hat{F}_{n_p}$$
$$+\ 2\frac{i}{\hbar}\sum_{i}^{n_p}\int d^3pd^3rV_{ep}(r-R_i)\sin\{\frac{\hbar}{2}\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i}\}\hat{F}_e(r,p)\hat{F}_{n_p} \qquad (19)$$

To make equations (18) and (19) more compact, we have used the notation

$$\hat{F}_{n_p}\equiv\hat{F}_{n_p}(R_1,...,R_{n_p},P_1,...,P_{n_p}) \qquad (20)$$

Equations (16) and (19) are the von Neumann equations for the electron-proton plasma. They constitute a complete description of the quantum dynamics of the system. If we were able to solve equation (16), and then substitute it into equation (19), we would have a complete description of the proton dynamics. The one-particle electron equation (16), is well known in various forms [57–64]. It still retains the presence of quantum diffraction effects through the non-locality of the right hand side. A manifestation of the BBGKY hierarchy is seen in the product of the one-particle phase space density operators on the right hand side of equation (16).

The $n_p$ proton von Neumann equation is an operator form generalization of the equation derived by Ludwig, Bonitz, Kahlert and Dufty [45]. It is the quantum operator analog of the proton Liouville equation. The first term on the right hand side of equation (18) is typical of the Liouville equation for a proton-only system. The second term is a manifestation of BBGKY and couples the $n_p$ and one-particle electron phase space density operators.

This term will be responsible for the coupling of the MD description of the protons to the continuum description of the electrons based on kinetic theory.

Equations (18) or (19) can be simplified even further if we make use of the observation that the plasma properties of hot dense matter relevant to burning ICF targets are such that the ions behave classically and the electrons exhibit both quantum and classical behavior. In order to capture the hybrid quantum-classical dynamics, we adopt the methods of Kapral and Cicotti [65]. They derived classical-quantum hybrid equations of motion for a quantum subsystem of light particles coupled to a classical bath of heavy particles. They performed a partial Wigner transform of the evolution equation of the density matrix for the whole system with respect to the classical system consisting of heavy particles. Scaling variables were then applied to the resulting equation followed by an expansion in the parameter $\mu = \sqrt{m/M}$ which is the square root of the ratio of the light (m) to heavy (M) mass particles. We do not simply perform an $\hbar$ expansion of equations (18) or (19) to extract the classical behavior since for Coulomb potentials each succeeding term in $\hbar$ becomes more singular.

We apply the scaling variables used in Kapral and Cicotti [65] to the proton von Neumann equation. If we define $\epsilon$ as some energy scale, then a thermal De Broglie length scale $\lambda$, time scale $\tau$, electron momentum $\Pi_e$ and proton momentum $\Pi_p$ can be defined by

$$\lambda = \hbar/\sqrt{(m_e\epsilon)}, \quad \tau = \frac{\hbar}{\epsilon}, \quad \Pi_e = \frac{\lambda m_e}{\tau}, \quad \Pi_p = \sqrt{m_e\epsilon} \Leftrightarrow \Pi_e = \sqrt{\frac{m_e}{m_p}}\Pi_p \equiv \mu\Pi_p \tag{21}$$

$$(\tilde{r},\tilde{p}) = \left(\frac{r}{\lambda},\frac{p}{\Pi_e}\right), \quad \left(\tilde{R},\tilde{P}\right) = \left(\frac{R}{\lambda},\frac{P}{\Pi_p}\right), \quad \tilde{t} = \frac{t}{\tau}, \quad \tilde{V_{ab}} = \frac{V_{ab}}{\epsilon}, \quad \hat{\Phi}_n = \hbar^{3n}\hat{F}_n \tag{22}$$

We obtain a transformed von Neumann equation given by,

$$\begin{aligned}\frac{\partial\hat{\Phi}_{n_p}}{\partial\tilde{t}} + \mu\sum_{i=1}^{n_p}\tilde{P}_i\cdot\nabla_{\tilde{R}_i}\hat{\Phi}_{n_p} \;=\; & 2\sum_{i<j}^{n_p}V_{pp}(\tilde{R}_i-\tilde{R}_j)\sin\{\frac{\mu}{2}\{\overleftarrow{\nabla}_{\tilde{R}_i}\cdot\overrightarrow{\nabla}_{\tilde{P}_i} + \overleftarrow{\nabla}_{\tilde{R}_j}\cdot\overrightarrow{\nabla}_{\tilde{P}_j}\}\}\hat{\Phi}_{n_p} \\ & + \; 2\sum_{i}^{n_p}\int d^3\tilde{p}d^3\tilde{r}\tilde{V}_{ep}(\tilde{r}-\tilde{R}_j)\sin\{\frac{\mu}{2}\overleftarrow{\nabla}_{\tilde{R}_i}\cdot\overrightarrow{\nabla}_{\tilde{P}_i}\}\hat{\Phi}_e(r,p)\hat{\Phi}_{n_p}\end{aligned} \tag{23}$$

Transforming back to the original variables and expanding in powers of $\mu$, we obtain

$$\frac{\partial \hat{F}_{n_p}}{\partial t} + \sum_{i=1}^{n_p} \frac{P_i}{m_p} \cdot \nabla_{R_i} \hat{F}_{n_p} = \sum_{i<j}^{n_p} V_{pp}(R_i - R_j)\{\overleftarrow{\nabla}_{R_i} \cdot \overrightarrow{\nabla}_{P_i} + \overleftarrow{\nabla}_{R_j} \cdot \overrightarrow{\nabla}_{P_j}\}\hat{F}_{n_p}$$
$$+ \sum_{i}^{n_p} \int d^3p d^3r V_{ep}(r - R_j) \overleftarrow{\nabla}_{R_i} \cdot \overrightarrow{\nabla}_{P_i} \hat{F}_e(r,p) \hat{F}_{n_p}$$
$$\equiv \hat{\mathcal{L}}_{CL} \hat{F}_{n_p} \tag{24}$$

where $\hat{\mathcal{L}}_{CL}$ is defined as the right hand side operator of equation (24).

Upon expansion in the ratio of electron to proton mass, the $n_p$ proton von Neumann equation takes on the classical form of the Liouville equation. There is a significant difference however, in that this equation is still in operator form with the "classical" Liouville operator $\hat{\mathcal{L}}_{CL}$ a functional of the electron phase space density operator. Equations (16) and (24) are in the desired form that will allow us to describe the plasma by coupling quantum kinetic and molecular dynamics descriptions.

## IV. WIGNER EQUATIONS AND CORRELATION FUNCTIONS

Based on the weak coupling approximation of the electron-electron and electron-proton interactions, expectation values of the electron and proton von Neumann equations can be written in closed form. In this section, we show how the BBGKY hierarchy can be reduced to a set of equations involving the one-particle electron distribution function, electron-electron and electron-proton two-particle correlation functions and the proton Liouville equation.

We begin by defining the quantum expectation value of an arbitrary n-particle operator $A_n(t)$ in the usual way

$$\langle \hat{A}_n(t) \rangle = Tr[\hat{A}_n(t)\rho_N] \tag{25}$$

where $N = n_p + n_e$, $\rho_N$ is the density operator for the plasma and n in equation (25) can take take on any value from 1 to N.

The expectation value of the phase space density operator is the Wigner distribution function. The one-particle electron and proton Wigner distribution functions $f_e(r,p)$ and $f_p(R,P)$ are defined as

$$\langle \hat{F}_e(r,p)\rangle = f_e(r,p), \qquad \langle \hat{F}_p(R,P)\rangle = f_p(R,P) \tag{26}$$

$$\langle \hat{F}_{n_p}(R_1:R_{n_p},P_1:P_{n_p})\rangle = f_{n_p}(R_1:R_{n_p},P_1:P_{n_p}) \equiv f_{n_p} \tag{27}$$

$$\int d^3P\, F_p(R,P) = N_P(R), \qquad \int d^3R\, d^3P\, F_P(R,P)) = n_p \tag{28}$$

$$\int d^3p f_e(r,p) = N_e(r), \qquad \int d^3r d^3p f_e(r,p) = n_e \tag{29}$$

Applying the expectation values to the one-electron and $n_p$-proton von Neumann equations, we obtain the set of Wigner equations for the one-particle electron and the $n_p$ proton distribution functions, viz.,

$$\begin{aligned}\frac{\partial f_e(r,p)}{\partial t} + \frac{p}{m_e}\cdot\nabla_r f_e(r,p) \;&=\; \frac{i}{\hbar}\sum_a \int \frac{d^3x}{(2\pi\hbar)^3} d^3q d^3r_a d^3s_a e^{i(p-q)\cdot x/\hbar} \\ &\times\; [V_{ea}(r-r_a+\frac{x}{2}) - V_{ea}(r-r_a-\frac{x}{2})] \\ &\times\; \langle \hat{F}_a(r_a,s_a)\hat{F}_e(r,q)\rangle \end{aligned} \tag{30}$$

$$\begin{aligned}\frac{\partial f_{n_p}}{\partial t} + \sum_{i=1}^{n_p}\frac{P_i}{m_p}\cdot\nabla_{R_i} f_{n_p} \;&=\; \sum_{i<j}^{n_p} V_{pp}(R_i-R_j)\{\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i} + \overleftarrow{\nabla}_{R_j}\cdot\overrightarrow{\nabla}_{P_j}\}\} f_{n_p} \\ &+\; \sum_i^{n_p}\int d^3p d^3r V_{ep}(r-R_i)\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i}\langle \hat{F}_e(r,p)\hat{F}_{n_p}\rangle \end{aligned} \tag{31}$$

We now recast the right hand side of equations (30) and (31) into a more illustrative form by defining a set of correlation functions $g_{ee}, g_{ep}$ and $g_{en_p}$ given by,

$$\langle \hat{F}_e(r_1,p_1)\hat{F}_e(r_2,p_2)\rangle = f_e(r_1,p_1)f_e(r_2,p_2) + g_{ee}(r_1,p_1;r_2,p_2) \tag{32}$$

$$\langle \hat{F}_e(r,p)\hat{F}_p(R,P)\rangle = f_e(r,p)f_p(R,P) + g_{ep}(r,p;R,P) \tag{33}$$

$$\langle \hat{F}_e(r,p)\hat{F}_{n_p}(R,P)\rangle = f_e(r,p)f_{n_p}(R_1:R_{n_p},P_1:P_{n_p}) + g_{en_p}(r,p;R_1:R_{n_p},P_1,P_{n_p}) \quad (34)$$

We will also have use later for the dimensionless correlation functions $h_{ab}(r_1,p_1;r_2,p_2)$, defined as

$$g_{ab}(r_1,p_1;r_2,p_2) = f_a(r_1,p_1)f_b(r_2,p_2)h_{ab}(r_1,p_1;r_2,p_2) \quad (35)$$

Upon substitution of equations (32) through (34) into equations (30) and (31), the one-particle electron Wigner and $n_p$ proton Wigner equations can be written

$$\begin{aligned}\frac{\partial f_e(r,p)}{\partial t} + \frac{p}{m_e}\cdot\nabla_r f_e(r,p) &= \frac{i}{\hbar}\sum_b \int d^{12}\Omega e^{i(p-\alpha)\cdot x/\hbar} \\ &\times [V_{eb}(r+x/2-s) - V_{eb}(r-x/2-s)] \\ &\times f_e(r,\alpha)f_b(s,\beta) \\ &+ \frac{i}{\hbar}\sum_b \int d^{12}\Omega e^{i(p-\alpha)\cdot x/\hbar} \\ &\times [V_{eb}(r+x/2-s) - V_{eb}(r-x/2-s)] \\ &\times g_{eb}(r,\alpha;s,\beta) \end{aligned} \quad (36)$$

$$\begin{aligned}\frac{\partial f_{n_p}}{\partial t} + \sum_{i=1}^{n_p}\frac{P_i}{m_p}\cdot\nabla_{R_i} f_{n_p} &= \sum_{i<j}^{n_p} V_{pp}(R_i-R_j)\{\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i} + \overleftarrow{\nabla}_{R_j}\cdot\overrightarrow{\nabla}_{P_j}\}\}f_{n_p} \\ &+ \sum_i^{n_p}\int d^3r V_{ep}(r-R_i)N_e(r)\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i} f_{n_p} \\ &+ \sum_i^{n_p}\int d^3r V_{ep}(r-R_i)N_e(r)\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i} \\ &\times g_{en_p}(r,p;R_1:R_{n_p},P_1:P_{n_p}) \end{aligned} \quad (37)$$

We have simplified the notation by writing

$$\int d^{12}\Omega\,(...) = \int \frac{d^3x}{(2\pi\hbar)^3} d^3s d^3\alpha d^3\beta\,(...) \quad (38)$$

As a consequence of BBGKY, these compact forms of the electron and many-particle proton Wigner equations demonstrate a need for the electron-electron, electron-proton, and

electron-$n_p$ proton correlation functions. We will show in the next section how the electron-$n_p$ proton correlation function can be written as a function of two-particle correlation functions. In this way, the coupled set of electron and proton Wigner equations (36) and (37) can be reduced to a description of the plasma based solely on single-particle distribution functions, two-particle correlation functions and an $n_p$ proton distribution function. The result is equations (46) and (47).

### A. Reduction of the Electron-Many Proton Correlation Function

Equation (37) requires further simplification due to the unwieldy correlation function $g_{en_p}(r,p;R_1 : R_{n_p}, P_1 : P_{n_p})$. In this form, the coupled set of equations is useless since it requires knowledge of how every proton correlates to a given electron. Guernsey has shown that if the pair-wise correlations are small, the expectation value of a product of the one-particle electron phase space density operator with the $n_p$ proton phase space density operator can be decomposed into a function of one-electron and one-proton distribution functions and their pair-wise correlations.

$$\begin{aligned}\langle \hat{F}_e(r,p)\hat{F}_{n_p}\rangle &= \langle \hat{F}_e(r,p)\rangle\langle \hat{F}_p(R_1,P_1)\rangle ... \langle \hat{F}_p(R_{n_p},P_{n_p})\rangle \\ &+ \sum_{i<j}^{n_p}\sum_{j=2}^{n_p} g_{pp}(R_i,P_i;R_j,P_j)\langle \hat{F}_e(r,p)\rangle \times \prod_{k=1,k\neq i,j}^{n_p}\langle \hat{F}_p(R_k,P_k)\rangle \\ &+ \sum_{i=1}^{n_p} g_{ep}(r,p;R_i,P_i)\times \prod_{k=1,k\neq i}^{n_p}\langle \hat{F}_p(R_k,P_k)\rangle \end{aligned} \tag{39}$$

This form is still not yet particularly useful as written, since it decomposes the left hand side into a function of one-particle functions. We would like the right hand side of equation (39) to contain the terms $\hat{F}_{n_p}$ since it preserves the Liouville form for the protons. The first two terms on the right hand side of equation (39) can be summed to obtain $\langle \hat{F}_e(r,p)\rangle\langle \hat{F}_{n_p}(R,P)\rangle$. Therefore,

$$\begin{aligned}\langle \hat{F}_e(r,p)\hat{F}_{n_p}\rangle &= f_e(r,p)f_{n_p} \\ &+ \sum_{i=1}^{n_p} g_{ep}(r,p;R_i,P_i)\times \prod_{k=1,k\neq i}^{n_p}\langle \hat{F}_p(R_k,P_k)\rangle \end{aligned} \tag{40}$$

The final step in writing the expectation value of the product of the one-electron and $n_p$ phase space density operators as a function of the expectation value of the n-proton phase space density operator is to note that to lowest order

$$\begin{aligned}
\langle \hat{F}_e(r,p)\hat{F}_{n_p}\rangle &= \langle \hat{F}_e(r,p)\rangle\langle \hat{F}_{n_p}\rangle \\
&+ \sum_{i=1}^{n_p} g_{ep}(r,p;R_i,P_i)\cdot \prod_{k=1,k\neq i}^{n_p}\langle \hat{F}_p(R_k,P_k)\rangle \\
&= \langle \hat{F}_e(r,p)\rangle\langle \hat{F}_{n_p}\rangle \\
&+ \langle \hat{F}_e(r,p)\rangle \sum_{i=1}^{n_p}\frac{g_{ep}(r,p;R_i,P_i)}{\langle \hat{F}_e(r,p)\rangle\langle \hat{F}_{n_p}\rangle}\langle \hat{F}_{n_p}\rangle \times \prod_{k=1,k\neq i}^{n_p}\langle \hat{F}_p(R_k,P_k)\rangle \\
&\approx f_e(r,p)f_{n_p} + f_e(r,p)f_{n_p}\sum_{i=1}^{n_p} h_{ep}(r,p;R_i,P_i) \qquad (41)
\end{aligned}$$

Upon substituting equation (41) into (37), we obtain

$$\begin{aligned}
\frac{\partial f_{n_p}}{\partial t} + \sum_{i=1}^{n_p}\frac{P_i}{m_p}\cdot\nabla_{R_i} f_{n_p} &= \sum_{i<j}^{n_p} V_{pp}(R_i-R_j)\{\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i} + \overleftarrow{\nabla}_{R_j}\cdot\overrightarrow{\nabla}_{P_j}\}\}f_{n_p} \\
&+ \sum_{i}^{n_p}\int d^3r V_{ep}(r-R_i)N_e(r)\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i} f_{n_p} \\
&+ \sum_{i}^{n_p}\int d^3p d^3r V_{ep}(r-R_i)f_e(r,p)\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i} \\
&\times [\sum_{j=1}^{n_p} h_{ep}(r,p;R_j;P_j)]f_{n_p} \qquad (42)
\end{aligned}$$

We will now simplify the last term in equation (42) by transforming the discrete sum over the various $h_{ep}$ contributions into an integral form. We begin by Fourier transforming the sum of the dimensionless correlation function $h_{ep}$.

$$\Phi(r,p) = \sum_{j=1}^{n_p} h_{ep}(r,p;R_j;P_j) = \sum_{j=1}^{n_p}\int \frac{d^3K}{(2\pi)^3}\frac{d^3Q}{(2\pi)^3} e^{iK\cdot R_j} e^{iQ\cdot P_j}\tilde{h}_{ep}(r,p;K,Q) \qquad (43)$$

If we switch the order of integration and summation in equation (43) and note that

$$\tilde{f}_p(K,Q) = \sum_{j=1}^{n_p} e^{iK\cdot R_j} e^{iQ\cdot P_j} \qquad (44)$$

is the Fourier transform of the one-particle proton distribution function $f_p(R,P)$, equation (43) can be written

$$\Phi(r,p) = \int d^3R' d^3P' f_p(R',P') h_{ep}(r,p;R',P') \tag{45}$$

### B. KTMD Equations and Two-Particle Correlation Functions

Using the results from the previous section, we are now in a position to describe the coupled electron-proton dynamics through a set of coupled Liouville and electron kinetic equations. Each equation will contain a term involving the electron-electron and electron-proton correlation functions. For now, we will leave the question as to how to treat the correlation functions to section IV. Substituting equation (45) into equation (42), and using the definition of $h_{ep}$ in equation (35), we obtain a simple form of the proton Liouville equation in terms of one-particle distribution and correlation functions.

$$\begin{aligned}
\frac{\partial f_{n_p}}{\partial t} + \sum_{i=1}^{n_p} \frac{P_i}{m_p} \cdot \nabla_{R_i} f_{n_p} \;=\; & \sum_{i<j}^{n_p} V_{pp}(R_i - R_j) \{ \overleftarrow{\nabla}_{R_i} \cdot \overrightarrow{\nabla}_{P_i} + \overleftarrow{\nabla}_{R_j} \cdot \overrightarrow{\nabla}_{P_j} \} f_{n_p} \\
& + \sum_{i}^{n_p} \int d^3r V_{ep}(r - R_i) N_e(r) \overleftarrow{\nabla}_{R_i} \cdot \overrightarrow{\nabla}_{P_i} f_{n_p} \\
& + \sum_{i}^{n_p} \int d^3p d^3r d^3R' d^3P' g_{ep}(r,p;R',P') V_{ep}(r - R_i) \overleftarrow{\nabla}_{R_i} \\
& \cdot \; \overrightarrow{\nabla}_{P_i} f_{n_p}
\end{aligned} \tag{46}$$

The electron dynamics are described by

$$\begin{aligned}
\frac{\partial f_e(r,p)}{\partial t} + \frac{p}{m_e} \cdot \nabla_r f_e(r,p) \;=\; & \frac{i}{\hbar} \sum_b \int d^{12}\Omega e^{i(p-\alpha)\cdot x/\hbar} \\
& \times \; [V_{eb}(r + x/2 - s) - V_{eb}(r - x/2 - s)] \\
& \times \; f_e(r,\alpha) f_b(s,\beta) \\
& + \; \frac{i}{\hbar} \sum_b \int d^{12}\Omega e^{i(p-\alpha)\cdot x/\hbar} \\
& \times \; [V_{eb}(r + x/2 - s) - V_{eb}(r - x/2 - s)] \\
& \times \; g_{eb}(r,\alpha;s,\beta)
\end{aligned} \tag{47}$$

,

We have repeated the electron kinetic equation for completeness. Assuming the electron-electron and electron-proton correlation functions are known (see the next section for three different variants), equations (46) and (47) are a coupled set of equations that describe the electron-proton plasma. Equations (46) and (47) are the desired KTMD equations. In the KTMD approach, the proton Liouville equation (46) describes the forces acting on each proton. The forces acting on the protons are dependent on classical Coulomb forces, one-particle electron distribution functions and the two-particle correlation functions. The first term on the right hand side of equation (46) describes the classical Coulomb interaction. The second term describes a mean-field contribution coming from the local electron density, and the third term is the contribution to the proton motion caused by electron-proton density fluctuations. Molecular dynamics provides the numerical method for solving this equation. The electron information comes from the kinetic equation described by equation (47). This equation is a multi-dimensional partial differential equation that can be solved by various techniques [66] depending on the choice of correlation function and other approximations. In the next section we describe three examples for the two-particle correlation functions appearing on the right hand side of equations and the physical situations in which they are applicable.

## V. PLASMA PROPERTIES AND THE CLOSED SET OF KTMD EQUATIONS FOR AN ELECTRON PROTON PLASMA

Based on the physical conditions of the plasma, constraints can be placed on the appropriate electron-proton and proton-proton correlation functions. The various options for the correlation functions will allow us to close the KTMD equations.

The simplest KTMD variant is based on a mean-field approximation that is appropriate for hot, diffuse, and weakly coupled plasmas. In this regime, the electron plasma frequency greatly exceeds all collision frequencies. In addition, the collisional mean-free path is much larger than the relevant system size. The mean field approximation cannot describe scattering processes and the potential energy of the system contains only mean field contributions. Relevant high-energy density plasmas for which KTMD in the mean-field approximation is applicable are Tokamak and some laser produced plasmas. Applications where this ap-

proach would not be valid include ion stopping in dense plasmas and collisional relaxation of electron-ion systems. Both of these applications require detailed knowledge of scattering processes. Ion diffusivity in diffuse plasmas is a good application for the mean-field approach.

The second KTMD variant is based on the Singwi-Tosi-Land-Sjolander (STLS) ansatz [67] where the electron-electron and electron-proton correlation functions are assumed static, and equal to the radial distribution function. It is well known that STLS is only valid near equilibrium. STLS extends mean-field theory to include equilibrium correlations. Even though STLS allows us to consider denser plasmas where the electrons can be strongly correlated but not far from equilibrium, our KTMD derivation assumes the electrons to be weakly correlated (see equation 41). This variant is illustrative of how non-zero correlations enter into the KTMD equations and modify the forces acting on the protons.

The third KTMD variant is the most complex and represents what we would call the full KTMD description of the plasma. The correlation functions are dynamic, position, momentum and time dependent, small, and their evolution is described by a set of quantum kinetic equations that arise from closing the BBGKY hierarchy at the three particle correlation level. Both the Landau and Lenard-Balescu equations are contained within this third variant of KTMD. The regime of weakly coupled and weakly collisional electrons is described by the full KTMD variant. Hence the relevant plasmas tend to be hot but more dense than the mean-field approximation would allow. These plasmas have their collision frequency comparable to or greater than the plasma frequency and the collisional mean-free path is smaller than the relevant system size. Inertial Confinement Fusion plasmas tend to belong to this category. We note that near equilibrium, the full KTMD variant is equivalent to STLS. Ion stopping in dense plasmas is a good application for this variant of KTMD.

### A. KTMD in the Mean-Field Limit

For hot and diffuse enough plasmas, the plasma frequency is much larger than the collision frequency. Physically, collective effects dominate over collisional processes and the plasma is nearly collisionless and we may take $g_{ee} = g_{ep} = g_{en_p} = 0$. we obtain a closed set of equations which define KTMD within the mean field approximation. Setting $g_{ee} = g_{ep} = g_{en_p} = 0$ in equations (46) and (47) yields a closed set of equations for the electron kinetic equation and

the proton particle dynamics. The picture is one where a continuous or fluid-like description of the electrons is coupled to a particle description of the ions. It is important to recognize that the proton-proton correlations need not be small and proton-proton correlations are included through the proton-proton Coulomb interaction in equation (35). The mean field KTMD equations are

$$\begin{aligned}\frac{\partial f_e(r,p)}{\partial t} + \frac{p}{m_e}\cdot\nabla_r f_e(r,p) &= \frac{i}{\hbar}\sum_b \int d^3\Omega e^{i(p-\alpha)\cdot x/\hbar} \\ &\times \left[V_{eb}(r+x/2-s) - V_{eb}(r-x/2-s)\right] \\ &\times f_e(r,\alpha) f_b(s,\beta)\end{aligned} \tag{48}$$

$$\begin{aligned}\frac{\partial f_{n_p}}{\partial t} + \sum_{i=1}^{n_p}\frac{P_i}{m_p}\cdot\nabla_{R_i} f_{n_p} &= \sum_{i<j}^{n_p} V_{pp}(R_i-R_j)\{\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i} + \overleftarrow{\nabla}_{R_j}\cdot\overrightarrow{\nabla}_{P_j}\} f_{n_p} \\ &+ \sum_{i}^{n_p}\int d^3r V_{ep}(r-R_i) N_e(r)\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i} f_{n_p}\end{aligned} \tag{49}$$

This set of equations defines completely the time evolution of the electron fluid and the classical point ions within the mean-field approximation. The evolution of the electron Wigner distribution function depends on two contributions. One is from the electron-electron interactions and the other is from the electron-proton interactions. The proton dynamics are described by a classical Liouville equation whose solution is obtained via MD. The forces acting on each proton can be isolated in equation (49). There is the usual classical proton-proton Coulomb force and an additional force due to the presence of the electron fluid. This latter force is the gradient of a mean potential defined by the convolution of the bare electron-proton Coulomb potential with the local time dependent electron density. The quantum nature of the electron-proton interaction is contained in the electron distribution function, but is classical otherwise (same as Vlasov). The effective potential is defined as,

$$V_{ep}^{eff} = \int d^3r V_{ep}(r-R_j) N_e(r). \tag{50}$$

This effective potential includes non-linear screening and is responsive to the time evolving electron dynamics. Therefore, it can be thought of as a generalization of the Yukawa potential. We note that the effective potential depends not on the six-dimensional distribution function but rather only on the zeroth order moment: the electron number density $N_e(r)$. As we will see, this observation is still valid beyond the mean-field approximation but only in the cases where the correlation functions are independent of momenta. Therefore, a quantum hydrodynamical description should be sufficient to describe the electron dynamics for certain cases within KTMD.

### B. KTMD and the Singwi-Tosi-Land-Sjolander (STLS) Ansatz

Equation (46) describes the many particle dynamics of the protons as they interact with the electron fluid and each other. In this section, we show that the last force term in equation (46) can be evaluated and takes on a simple form if the STLS ([67–69]) ansatz is invoked. We assume that the two-particle correlation function is independent of momentum and it is only a function of the coordinate difference. As noted in the introduction to this section, the STLS ansatz extends the mean-field approximation by including equilibrium electron-electron and electron-proton correlations. The STLS ansatz is in general valid for large correlations but where the system is near equilibrium. However, our KTMD derivation assumed the electrons to be weakly correlated (see equation (41)). Therefore, the STLS variation of KTMD presented here is valid for weakly correlated electrons only. In spite of this, the STLS variant of KTMD allows one to move beyond mean-field.

$$\langle \hat{F}_e(r,p)\hat{F}_p(R,P)\rangle = f_e(r,p)f_p(R,P)(1+h_{ep}(r,p;R,P)) \approx f_e(r,p)f_p(R,P)\xi_{ep}(r-R) \quad (51)$$

The quantity $\xi_{ep}(r-R)$ is the radial distribution function which describes the likelihood that there is an electron at position r given that there is a proton at position R. The radial distribution function is related to the static structure factor $\tilde{S}_{ep}(k)$ in the usual way [70],

$$\tilde{S}_{ep}(k) = \sqrt{n_e n_p}\int d^3 r e^{ik\cdot r}(\xi_{ep}(r)-1) \quad (52)$$

In the STLS ansatz, the relationship between the radial distribution function, static structure factor, and dielectric function, provide a set of equations that can be solved self

consistently [67, 71]. From equations (51) and (52)we can write,

$$\tilde{h}_{ep}(k) = \tilde{\xi}_{ep}(k) - (2\pi)^3\delta^3(k) = S_{ep}(k)/\sqrt{n_e n_p} \tag{53}$$

To simplify the last term in equation (42), we write,

$$\Phi(r) = \sum_{i=1}^{n_p} h_{ep}(r, p; R_i; P_i) \approx \sum_{i=1}^{n_p} h_{ep}(r - R_i) = \sum_{i=1}^{n_p} \int \frac{d^3k}{(2\pi)^3} e^{ik\cdot(r-R_i)} \tilde{h}_{ep}(k) \tag{54}$$

If we switch the order of the integration and summation in equation (54) and note that the proton number density $N_p(r)$ and its Fourier transform $\tilde{N}_p(k)$ can be written

$$N_p(r) = \sum_{i=1}^{n_p} \delta^3(R - R_i) \Leftrightarrow \tilde{N}_p(k) = \sum_{i=1}^{n_p} e^{-ik\cdot R_i} \tag{55}$$

Using equation (55), equation (54) can be written,

$$\Phi(r) = \int \frac{d^3k}{(2\pi)^3} e^{ik\cdot r} \tilde{N}_p(k)\tilde{h}_{ep}(k) = \int \frac{d^3k}{(2\pi)^3} e^{ik\cdot r} \tilde{N}_p(k) \frac{\tilde{S}_{ep}(k)}{\sqrt{n_e n_p}} = \int d^3s \frac{S_{ep}(r-s)}{\sqrt{n_e n_p}} N_p(s) \tag{56}$$

Substituting equation (56) into the proton Liouville equation (42) we obtain a form amenable to a molecular dynamics code,

$$\begin{aligned} \frac{\partial f_{n_p}}{\partial t} + \sum_{i=1}^{n_p} \frac{P_i}{m_p} \cdot \nabla_{R_i} f_{n_p} \;=\; & \sum_{i<j}^{n_p} V_{pp}(R_i - R_j)\{\overleftarrow{\nabla}_{R_i} \cdot \overrightarrow{\nabla}_{P_i} + \overleftarrow{\nabla}_{R_j} \cdot \overrightarrow{\nabla}_{P_j}\}\} f_{n_p} \\ & + \sum_{i}^{n_p} \int d^3r V_{ep}(r - R_i) N_e(r) \overleftarrow{\nabla}_{R_i} \cdot \overrightarrow{\nabla}_{P_i} f_{n_p} \\ & + \sum_{i}^{n_p} \int d^3p d^3r \Phi(r) V_{ep}(r - R_i) f_e(r, p) \\ & \times \overleftarrow{\nabla}_{R_i} \cdot \overrightarrow{\nabla}_{P_i} f_{n_p} \end{aligned} \tag{57}$$

From equation (57), it is clear there are three force terms acting on each proton. The first is the pure Coulomb force, the second the mean-field force, and the third is a consequence of the equilibrium density fluctuations within the STLS ansatz,

$$F_i = \overrightarrow{\nabla}_{R_i} [\int d^3r d^3s \frac{S_{ep}(r-s)}{\sqrt{n_e n_p}} N_p(s) N_e(r) V_{ep}(r - R_i)] \tag{58}$$

The electron kinetic equation that couples to equation (57) using the STLS approximation is

$$\begin{aligned}\frac{\partial f_e(r,p)}{\partial t} + \frac{p}{m_e}\cdot\nabla_r f_e(r,p) \;&=\; \frac{i}{\hbar}\sum_b \int d^{12}\Omega e^{i(p-\alpha)\cdot x/\hbar} \\ &\times\; [V_{eb}(r+x/2-s) - V_{eb}(r-x/2-s)] \\ &\times\; f_e(r,\alpha) f_b(s,\beta)\xi_{eb}(r-s)\end{aligned} \tag{59}$$

As mentioned at the beginning of this section, self consistency requirements between the dielectric functions, static structure factor, and radial distribution function determines the form of $S_{ep}$ in equation (57). The STLS ansatz allows for correlated electrons that are near equilibrium. Hence, it is another extension of Yukawa to correlated but near equilibrium systems. Whereas the mean-field variant of KTMD would not be applicable to electron-ion relaxation where the electron temperature is changing, KTMD with the STLS ansatz for the correlation functions would be applicable to systems where the ions are relaxing to a fixed electron temperature.

### C. Particle Collisions and Weak Correlations

The mean-field approximation is a useful illustration of the KTMD approach but for many applications in HDM, such as electron-ion relaxation and ion stopping, scattering processes are important. Therefore, the effects from non-zero correlations is needed. The STLS approach includes correlations but relies on the equilibrium approximation. In this section, we describe a version of KTMD that fully realizes the concept of a quantum kinetic equation for the electrons and MD for the protons that was described in the introduction. The quantum kinetic equations consist of a set of coupled kinetic equations for the one-particle distribution function and the two-particle electron-electron and electron-proton correlation functions. For example, equations (46) and (47) require a description of the electron and proton two-particle correlation functions. In particular, we need a time evolution equation for $g_{ee}$ and $g_{ep}$. The set of closed equations for $f_e(r,p), g_{ee}$ and $g_{ep}$ has been written down by Guernsey [60], Klimontovich [57], Balescu [62], Bonitz [58] and many others. We summarize the main steps of the derivation to arrive at a form that is amenable to computation.

In order to derive an equation for the correlation functions $g_{ee}$ and $g_{ep}$, we start with the expectation value of a product of one-particle density operators defined by equation (12),

$$G_{ea}(u,v;w,x) = \langle \hat{\Delta}_e(u,v)\hat{\Delta}_a(w,x)\rangle - \langle \hat{\Delta}_e(u,v)\rangle\langle \hat{\Delta}_a(w,x)\rangle \tag{60}$$

The index a can be "e" or "p". We have introduced a transformed correlation function $G_{ea}$ which is related to the correlation functions defined in equations (32) and (33).

$$\begin{aligned} g_{ea}(r,p;r_a,p_a) &= \int \frac{d^3x}{(2\pi\hbar)^3}\int \frac{d^3y}{(2\pi\hbar)^3} e^{ip\cdot x}e^{ip_a\cdot y} \\ &\times\ G_{ea}(r+x/2, r-x/2; r_a+y/2, r_a-y/2) \end{aligned} \tag{61}$$

For HDM, the electron-electron and electron-proton correlations are weak. We therefore truncate the triple product of one-particle density operators as [64],

$$\begin{aligned} \langle \Delta_a(u,v)\Delta_b(w,x)\Delta_c(s,t)\rangle &= \langle \Delta_a(u,v)\rangle\langle \Delta_b(w,x)\rangle\langle \Delta_c(s,t)\rangle \\ &+ \langle \Delta_a(u,v)\rangle G_{bc}(w,x;s,t) \\ &+ \langle \Delta_b(w,x)\rangle G_{ac}(u,v;s,t) \\ &+ \langle \Delta_a(s,t)\rangle G_{ab}(u,v;w,x) \end{aligned} \tag{62}$$

In order to obtain a kinetic equation for the $G_{ea}$ correlation functions, we differentiate equation (60) with respect to time and use equations (5-9) along with the truncation equation (62) to obtain

$$\begin{aligned} \frac{\partial}{\partial t}G_{ea}(u,v;w,x) &= -i\hbar\left(\frac{1}{2m_e}[{\nabla_u}^2 - {\nabla_v}^2] + \frac{1}{2m_a}[{\nabla_w}^2 - {\nabla_x}^2]\right)G_{ea}(u,v;w,x) \\ &+ \frac{i}{\hbar}\sum_b \int d^3s[V_{eb}(u-s) - V_{eb}(v-s) \\ &+ V_{eb}(w-s) - V_{eb}(x-s)]\Delta_b(s,s)G_{ea}(u,v;w,x) \\ &+ \frac{i}{\hbar}\sum_b \int d^3s[V_{eb}(u-s) - V_{eb}(v-s)]\Delta_e(u,v)G_{ba}(s,s;w,x) \\ &+ \frac{i}{\hbar}\sum_b \int d^3s[V_{eb}(w-s) - V_{eb}(x-s)]\Delta_a(w,x)G_{eb}(u,v;s,s) \end{aligned} \tag{63}$$

Applying the Wigner transform, and again using the notation

$$\int d^{12}\Omega = \int \frac{d^3x}{(2\pi\hbar)^3} d^3s d^3\alpha d^3\beta \tag{64}$$

The required equations for $g_{ee}$ and $g_{ep}$ are obtained,

$$\begin{aligned}
\frac{\partial}{\partial t} g_{ea}(r,p;r_a,p_a) &= [\frac{p}{2m_e}\cdot\nabla_r + \frac{p_a}{2m_a}\cdot\nabla_{r_a}] g_{ea}(r,p;r_a,p_a) \\
&+ \frac{i}{\hbar}\sum_b \int d^{12}\Omega e^{(p-\alpha)\cdot x/\hbar}[V_{eb}(r+x/2-s) - V_{eb}(r-x/2-s)] \\
&\times f_b(r_b,\beta) g_{ea}(r,\alpha;r_a,p_a) \\
&+ \frac{i}{\hbar}\sum_b \int d^{12}\Omega e^{(p_a-\alpha)\cdot x/\hbar}[V_{eb}(r_a+x/2-s) - V_{eb}(r_a-x/2-s)] \\
&\times f_b(r_b,\beta) g_{ea}(r,p;r_a,\alpha) \\
&+ \frac{i}{\hbar}\sum_b \int d^{12}\Omega e^{(p-\beta)\cdot x/\hbar}[V_{eb}(r+x/2-s) - V_{eb}(r-x/2-s)] \\
&\times f_e(r,\beta) g_{eb}(r_e,\alpha;r_a,p_a) \\
&+ \frac{i}{\hbar}\sum_b \int d^{12}\Omega e^{(p_a-\beta)\cdot x/\hbar}[V_{eb}(r_a+x/2-s) - V_{eb}(r_a-x/2-s)] \\
&\times f_a(r_a,\beta) g_{eb}(r,p;r_a,\alpha)
\end{aligned} \tag{65}$$

Equation (65) represents a pair of equations, a,b equals "e" or "p". For a,b=e, $r_b = r'$ and $p_b = p'$. While for a,b=p, $r_b = R$ and $p_b = P$. The pair of equations (65) requires knowledge of the one-particle electron and proton distribution functions. The former comes from equation (47) while the latter comes from the proton MD simulation. Equations (47) and (65) are the most general description of the non-degenerate electron dynamics, assuming weak electron-electron and electron-proton correlations ([58], [64], [60] ). Given the one-particle proton distribution function which comes from the MD simulation, it describes an inhomogeneous, time dependent, non-local (due to quantum diffraction), non-Markovian, dynamically screened electron plasma. No assumption is made concerning the relative time scales of correlations versus one-particle distributions functions (Bogoliubov hypothesis). In the limit of spatial homogeneity and assuming the Bogoliubov hypothesis, this set of equations becomes the quantum Lenard-Balescu equation for non-degenerate electrons, [60]. If a further simplification is invoked, namely static screening, then this equation becomes the quantum Landau equation. Thus KTMD allows flexibility in that, given an MD calculation of the one-particle proton distribution function, the electron dynamics can be described by any number of approximations (including static) to the full electron kinetic equation.

In summary, with equation (65), the closed set of KTMD equations are,

$$\begin{aligned}\frac{\partial f_e(r_e,p_e)}{\partial t}+\frac{p_e}{m_e}\cdot\nabla_{r_e}f_e(r_e,p_e) &= \frac{i}{\hbar}\sum_b\int d^{12}\Omega_b\, e^{i(p_e-\alpha)\cdot x_e/\hbar}\\ &\times\ [V_{eb}(r_e+x_e/2-s_b)-V_{eb}(r_e-x_e/2-s_b)]\\ &\times\ f_e(r_e,\alpha)f_b(s_b,\beta_b)\\ &+\ \frac{i}{\hbar}\sum_b\int d^{12}\Omega_b\, e^{i(p_e-\alpha)\cdot x_e/\hbar}\\ &\times\ [V_{eb}(r_e+x_e/2-s_b)-V_{eb}(r_e-x_e/2-s_b)]\\ &\times\ g_{eb}(r_e,\alpha;s_b,\beta_b) \qquad (66)\end{aligned}$$

$$\begin{aligned}\frac{\partial f_{n_p}}{\partial t}+\sum_{i=1}^{n_p}\frac{P_i}{m_p}\cdot\nabla_{R_i}f_{n_p} &= \sum_{i<j}^{n_p}V_{pp}(R_i-R_j)\{\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i}+\overleftarrow{\nabla}_{R_j}\cdot\overrightarrow{\nabla}_{P_j}\}\}f_{n_p}\\ &+\ \sum_i^{n_p}\int d^3rV_{ep}(r-R_i)N_e(r)\overleftarrow{\nabla}_{R_i}\cdot\overrightarrow{\nabla}_{P_i}f_{n_p}\\ &+\ \sum_i^{n_p}\int d^3p\,d^3r\,d^3R'\,d^3P'g_{ep}(r,p;R',P')V_{ep}(r-R_i)\overleftarrow{\nabla}_{R_i}\\ &\times\ \overrightarrow{\nabla}_{P_i}f_{n_p} \qquad (67)\end{aligned}$$

In this full realization of KTMD, we first note that all quantum effects associated with the electron-electron and electron-proton interactions are contained in the one-particle electron distribution function and the electron-electron and electron-proton correlation functions. Second, beyond the classical proton-proton Coulomb forces, the proton dynamics are modified by a dynamical mean-field force and a force term due to the correlations. The force term due to correlations introduces a momentum dependence into the effective potential between ions.

## VI. CONCLUSION

A hybrid method based on coupling a continuum treatment of electrons using quantum kinetic theory with a molecular dynamics treatment of the ions has been derived from a first principles approach based on the many-body von Neumann equations. Even though the derivation has been done for a fully ionized electron-proton plasma, the remarks given

here will refer to the more general case of electrons and ions in a fully ionized plasma. KTMD is flexible in its treatment of the electrons. It has the advantage that the quantum kinetic equation describing the electron dynamics can take on a variety of forms based on the electron plasma physical conditions and the approximations one wants to invoke. At the basic level, the quantum kinetic equation for the electrons (43) is the typical Wigner equation first derived by Balescu [59] and Guernsey [60]. However, depending on the application, this kinetic equation can be approximated by the quantum Lenard-Balescu [59, 60, 64] or quantum Landau equations[72].

In the introduction, the classical-quantum dual nature of electrons and their treatment with QSP was discussed. The electrons in KTMD are treated as a continuous medium or fluid, which interacts with the ions. The electron plasma spatial and temporal evolution is described by a set of coupled quantum kinetic equations for the one-particle and two-particle correlation functions. Within KTMD, the ions are classical point particles modeled with MD. The ions evolve according to the classical Newton equations but with a set of effective ion-ion potentials dependent on the local electron density and electron-proton correlations. Unlike QSP, the effective ion-ion interaction is fully consistent with the dynamics of the system.

Finally, it should be noted that both the mean-field and STLS variations of KTMD involve only the electron density. Another option that will be considered in the future is to solve a set of moment equations derived from the electron quantum kinetic equations. The moment equations are essentially a quantum hydrodynamic description of the electron plasma. The utility of this approach is that a coupled set of field variables (density, momentum and energy) obeying a set of hydrodynamic equations should be easier to solve than a coupled set of kinetic equations.

## VII. ACKNOWLEDGMENTS

F.R.G. wishes to thank Michael Bontiz, Jerome Daligault, Jim Dufty, Michael Desjarlais and Sam Trickey for many useful and enlightening conversations. F.R.G and M.S.M also wish to thank Russ Caflisch and Christian Ratsch for their warm hospitality during the Institute for Pure and Applied Mathematics (UCLA) Long Program on high-energy-density physics, where portions of this work were discussed and completed. This work was performed under

the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344 and Los Alamos National Security, LLC. (LANS), operator of the Los Alamos National Laboratory under Contract No. DE-AC52-06NA25396 with the U.S. Department of Energy. This work was funded by the Laboratory Directed Research and Development Program at LLNL under project tracking code 09-SI-011.

[1] C. A. Haynam et al. National Ignition Facility laser performance status. *Applied Optics*, 46: 3276, 2007.

[2] P. Emma et al. First lasing and operation of an angstrom-wavelength free-electron laser. *Nature Photonics*, 4:641, 2010.

[3] F. R. Graziani et al. Large-scale molecular dynamics simulations of dense plasmas: The cimarron project. *High Energy Density Physics*, 8:105, 2012.

[4] B. J. Alder and T. E. Wainwright. Studies in molecular dynamics. i. general method. *The Journal of Chemical Physics*, 31(2):459–466, 1959.

[5] E. L. Pollock and J. Glosli. Comments on P3M, FMM, and the Ewald method for large periodic coulombic systems. *Computer Physics Communications*, 95:93–110, 1996.

[6] M. Griebel, S. Knapek, and G. Zumbusch. *Numerical Simulation in Molecular Dynamics*. Springer-Verlag, Berlin, Germany, first edition, 2007.

[7] M. Tuckerman. *Statistical Mechanics: Theory and Molecular Simulation*. Oxford University Press, New York, first edition, 2010.

[8] M.S. Murillo. Strongly Coupled Plasmas and High Energy Density Matter. *Physics of Plasmas*, 11:2964, 2004.

[9] R.P. Drake. *High-Energy Density Physics*. Springer-Verlag, Berlin, first edition, 2006.

[10] S. Azteni and J. Meyer-ter-Vehn. *The Physics of Inertial Fusion*. Clarendon Press, Oxford, 2nd ed. edition, 2004.

[11] S. Ichimaru. Strongly Coupled Plasmas. *Physics Reports*, 54:1017, 1982.

[12] S. Ichimaru, H. Iyetomi, and S. Tanaka. Statistical Physics of Dense Plasmas: Thermodynamics, Transport Coefficients and Dynamic Correlations. *Physics Reports*, 149:91, 1987.

[13] J. Myatt, W. Theobald, J. A. Delettrez, C. Stoeckl, and M. Storm. High-Intensity laser interactions with mass-limited solid targets and implications for fast-ignition experiments on OMEGA EP. *Physics of Plasmas*, 14:056301, 2007.

[14] R. P. J. Town et al. Analysis of the National Ignition Facility Ignition Hohlraum Energetics Experiments. *Physics of Plasmas*, 18:056302, 2011.

[15] J. N. Glosli et al. Molecular dynamics simulations of temperature equilibration in dense hydrogen. *Phys. Rev. E*, 78:025401(R), 2008.

[16] G. Dimonte and J. Daligault. Molecular-dynamics simulations of electron-ion temperature relaxation in a classical coulomb plasma. *Phys. Rev. Lett.*, 101:135001, 2008.

[17] L. X. Benedict et al. Molecular dynamics simulations of electron-ion temperature equilibration in an $SF_6$ plasma. *Phys. Rev. Lett.*, 102:205004, 2009.

[18] J. Daligault and G. Dimonte. Correlation effects on the temperature-relaxation rates in dense plasmas. *Phys. Rev. E*, 79:056403, 2009.

[19] L. X. Benedict et al. Molecular dynamics simulations and generalized Lenard-Balescu calculations of electron-ion temperature equilibration in plasmas. *Phys. Rev. E*, 86:046406, 2012.

[20] M. S. Murillo and M. W. C. Dharma-wardana. Temperature relaxation in dense hydrogen. *Physical Review Letters*, 100:205005, 2008.

[21] P . E. Grabowski, M. P. Surh, D. F. Richards, A. B. Langdon, F. R. Graziani, and M. S. Murillo. Large-scale molecular dynamics studies of classical stopping power. *submitted to Phys. Rev. Lett.*, 2013.

[22] K. Mussack and W. Dappen. Dynamic screening screening for solar p-p reaction rates. *ApJ.*, 729:96, 2011.

[23] T. Haxhimali and R. Rudd. Estimation of diffusivity in binary yukawa mixtures in the warm dense matter regime. *submitted to Phy. Rev. E*, 2013.

[24] M.P. Surh, III Barbee, T.W., and L.H. Yang. First principles molecular dynamics of dense plasmas. *Phys. Rev. Lett.*, 86:5958–61, 2001.

[25] G. Salin and J.-M. Caillol. Equilibrium molecular dynamics simulations of the transport coefficients of the Yukawa one component plasma. *Physics of Plasmas*, 10:1220, 2003.

[26] J.-P. Hansen and I. R. McDonald. Thermal relaxation in a strongly coupled 2-temperature plasma. *Physics Letters A*, 97:42, 1983.

[27] U. Reimann and C. Toepffer. Temperature relaxation in a strongly coupled plasma. In *5th International Workshop on Atomic Physics for Ion Driven Fusion, Jan 29–Feb 02, 1990 Schliersee, Fed Rep Ger*, volume 8, page 771, 1990.

[28] G. Kelbg. Theorie des quanten-plasmas. *Annalen der Physik*, 12:219, 1963.

[29] T. Dunn and A. A. Broyles. Method for determining the thermodynamic properties of the quantum electron gas. *Phys. Rev.*, 157:156, 1967.

[30] C. Deutsch, M. M. Gombert, and H. Minoo. Classical modelization of symmetry effects in the dense high-temperature electron gas. *Physics Letters A*, 66:381, 1978.

[31] C. Deutsch, M. M. Gombert, and H. Minoo. Classical modelization of symmetry effects in the dense high-temperature electron gas: Errata. *Physics Letters A*, 72:481, 1979.

[32] F. Lado. Effective potential description of the quantum ideal gases. *Journal of Chemical Physics*, 47:5369, 1967.

[33] H. Minoo, M. M. Gombert, and C. Deutsch. Temperature-dependent coulomb interactions in hydrogenic systems. *Phys. Rev. A*, 23(2):924, Feb 1981.

[34] E. L. Pollock. Properties and computation of the coulomb pair density matrix. *Computer Physics Communications*, 52(1):49, 1988.

[35] B. Talin, A. Calisti, E. Dufour, and J. Dufty. Classical dynamics of electrons surrounding ions in hot and dense plasmas-related topics. *JQSRT*, 71:729, 2001.

[36] A.V. Filinov, M. Bonitz, and W. Ebeling. Improved Kelbg potential for correlated Coulomb systems. In *Journal of Physics A (Mathematical and General)*, volume 36, page 5957. IOP Publishing, 2003. International Conference on Strongly Coupled Coulomb Systems (SCCS), 2-6 September 2002, Santa Fe, NM, USA.

[37] A. V. Filinov, V. O. Golubnychiy, M. Bonitz, W. Ebeling, and J. W. Dufty. Temperature-dependent quantum pair potentials and their application to dense partially ionized hydrogen plasmas. *Phys. Rev. E*, 70(4):046411, Oct 2004.

[38] C. Jones and M. S. Murillo. Analysis of semi-classical potentials for molecular dynamics and monte carlo simulations of warm dense matter. *High Energy Density Physics*, 3(3-4):379, 2007.

[39] H. Feldmeier. Fermionic molecular dynamics. *Nuc. Phys. A*, 515:147, 1990.

[40] D. Klakow, C. Toepffer, and P.-G. Reinhard. Semiclassical molecular dynamics for strongly coupled coulomb systems. *J. Chem Phys.*, 101:10766, 1994.

[41] H. Feldmeier and J. Schnack. Molecular dynamics for fermions. *Rev. Mod. Phys.*, 72:655, 2000.

[42] M. Knaup, P.-G. Reinhard, C. Toepffer, and G. Zwicknagel. Wave packet molecular dynamics simulations of warm dense hydrogen. *J. Phys. A: Math. Gen.*, 36:6165, 2003.

[43] P. Grabowski et al. Wave packet spreading and localization in electron-nuclear scattering. *Phys. Rev. E*, 36:6165, 2013.

[44] D. Michta, M. P. Surh, and F. R. Graziani. Kinetic theory molecular dynamics: Numerical considerations. *High Energy Density Physics, in press*, 9:696, 2013.

[45] P. Ludwig, M. Bonitz, H. Kahlert, and J. W. Dufty. Dynamics of strongly correlated ions in a

partially ionized quantum plasma. *Journal of Physics: Conference Series*, 220:012003, 2010.

[46] J.P. Hansen and I.R. McDonald. Microscopic simulation of a strongly coupled hydrogen plasma. *Physical Review A (General Physics)*, 23:2041, 1981.

[47] B. Bernu and Hansen J.-P. Thermal-conductivity of a strongly coupled hydrogen plasma. *Physical Review Letters*, 48:1375, 1982.

[48] H. Totsuji, T. Kishimoto, and C. Totsuji. Structure of confined yukawa system (dusty plasma). *Phys. Rev. Lett.*, 78:3113, 1997.

[49] T. Saigo and S. Hamaguchi. Shear viscosity of strongly coupled yukawa systems. *Phys. Plasmas*, 9:1210, 2002.

[50] M. Murillo. Viscosity estimates of liquid metals and warm dense matter using the yukawa reference system. *High Energy Density Phys.*, 4:49, 2008.

[51] G. Kresse and J. Hafner. Ab initio molecular dynamics for liquid metals. *Phys. Rev. B*, 47: 558, 1994.

[52] E. Smargiassi and P. Madden. Orbital-free kinetic-energy functionals for first-principles molecular dynamics. *Phys. Rev. B*, 49:5220, 1994.

[53] M. P. Desjarlais, J. D. Kress, and L. A. Collins. Electrical conductivity for warm, dense aluminum plasmas and liquids. *Phys. Rev. E*, 66:025401, 2002.

[54] F. Lambert, V. Recoules, A. Decoster, J. Clrouin, and M. Desjarlais. On the transport coefficients of hydrogen in the inertial confinement fusion regime. *Phys. Plasmas*, 18:056306, 2011.

[55] A. Correa, J. Kohanoff, E. Artacho, D. Sanchez-Portal, and A. Caro. Nonadiabatic forces in ion-solid interactions: The initial stages of radiation damage. *Phys. Rev. Lett.*, 108:213201, 2012.

[56] A. Schleife, E. Draeger, Y. Kanai, and A. Correa1. Plane-wave pseudopotential implementation of explicit integrators for time-dependent kohn-sham equations in large-scale simulations. *J. Chem. Phys.*, 137:22A546, 2012.

[57] Y. Klimontovich. *Statistical Phyiscs*. Harwood Academic Publishers, London, first edition, 1986.

[58] M. Bonitz. *Quantum Kinetic Theory*. B. G. Teubner, Stuttgart. Germany, first edition, 1998.

[59] R. Balescu. Approach to equilibrium of a quantum plasma. *Phys. Fluids*, 4:94, 1962.

[60] R. I. Guernsey. Kinetic theory of quantum plasmas. *Phys. Rev.*, 127:1446, 1962.

[61] R. Balescu. *Statistical Mechanics of Charged Particles.* John Wiley and Sons, London, New York, Sydney, first edition, 1963.

[62] R. Balescu. *Equlibrium and Nonequilibrium Statistical Mechanics.* Krieger Publishing Company, Malabar, Florida, reprint edition, 1991.

[63] V. V. Belyi, Yu. A. Kukharenko, and J. Wallenborn. Pair correlation function and nonlinear kinetic equation for a spatially uniform polarizable nonideal plasma. *Phys. Rev. Lett.*, 76: 3554, 1996.

[64] V. V. Belyi and Yu. A. Kukharenko. The balescu kinetic equation with exchange interaction. *J. Stat. Mech.*, 6:06002, 2009.

[65] R. Kapral and G. Ciccotti. Mixed quantum-classical dynamics. *J. Chem. Phys.*, 110(18):8919, 1999.

[66] C. A. J. Fletcher. *Computational Techniques for Fluid Dynamics Volume 1.* Springer-Verlag, Berlin, Germany, second edition, 1997.

[67] K. S. Singwi, M. P. Tosi, R. H. Land, and A. Sjolander. Electron correlations at metallic densities. *Phys. Rev.*, 176(2):589, 1968.

[68] R. L. Liboff. Generalized bogoliubov hypothesis for dense fluids. *Phys. Rev. E*, 31:1883, 1985.

[69] R. L. Liboff and G. K. Schenter. Criterion for phase change in terms of oscillation of the pair distribution function. *J. Chem. Phys.*, 84:7005, 1986.

[70] J.-P. Hansen and I. R. McDonald. *Theory of Simple Liquids.* Academic Press, Amsterdam, 3rd ed. edition, 2006.

[71] S. Ichimaru, S. Mitake, S. Tanaka, and X.-Z. Yan. Theory of interparticle correlations in dense, high-temperature plasmas. *Phys. Rev. A*, 32(3):1768, 1985.

[72] D. Kremp, T. Bornath, M. Schlanges, and W-D. Kraeft. *Quantum Statistics of Nonideal Plasmas.* Springer-Verlag, Berlin, first edition, 2005.