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Molecular dynamics studies of electron-ion temperature equilibration in hydrogen plasmas within the coupled-mode regime

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Abstract

We use classical molecular dynamics (MD) to study electron-ion temperature equilibration in two-component plasmas in regimes for which the presence of coupled collective modes has been predicted to substantively reduce the equilibration rate. Guided by previous kinetic theory work, we examine hydrogen plasmas at a density of $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, and $10^7 \text{ K} < T_e < 10^9 \text{ K}$. The non-equilibrium classical MD simulations are performed with inter-particle interactions modeled by quantum statistical potentials (QSPs). Our MD results indicate: 1. a large effect from time-varying potential energy, which we quantify by appealing to an adiabatic 2-temperature equation of state, and 2. a notable deviation in the energy equilibration rate when compared to calculations from classical Lenard-Balescu theory including the QSPs. In particular, it is shown that the energy equilibration rates from MD are more similar to those of the theory when coupled modes are *neglected*. We suggest possible reasons for this surprising result, and propose directions of further research along these lines.

I. INTRODUCTION

The phenomenon of electron-ion temperature equilibration is important for the study of inertial confinement fusion (ICF), since the asymmetric manner in which α -particle fusion products deposit energy naturally creates such a temperature-split (T -split) at certain key regions and times within an imploding capsule [1, 2]. While much of the ICF capsule material is DT, the 50-50 mixture of deuterium and tritium, the presence of an outer, so-called, ablator layer in such capsules creates the possibility of admixture of higher- Z (ionic charge) elements (C, Be, and/or dopants like Ge and W) into the DT fuel. This in turn introduces significant complexities for the theoretical description of the T -equilibration process, because such ions serve to make the plasma more strongly-coupled. Since modeling of electron-ion T -equilibration is a necessary part of integrated simulations of ICF, the problems of T -equilibration in both low- Z and high- Z /low- Z mixtures are therefore of concern [3].

Recent work on T -equilibration has involved both kinetic theory of various types [4–8], and MD simulations with like-charges and the pure Coulomb interaction [8, 9] as well as opposite-charge simulations with quantum statistical potentials (QSPs) to mock up the effects of quantum diffraction [10–14]. All but a few of these studies [7, 11, 14] confined themselves to pure hydrogen (or the proton-positron system [8, 9]). Even for H, the demonstration of agreement between MD results and kinetic theory took some time [10–13], in part because the effect of using the QSPs changes the result slightly even in weak coupling [11]. For the classical like-charge proton-positron system, demonstration of agreement was somewhat more straightforward, motivating a clear connection between the classic Landau-Spitzer equilibration rate [15] derived from the Fokker-Planck equation, and a generalized Lenard-Balescu [16] scheme in which both dynamical screening and 2-body static correlations (in the form of local field corrections) are included [8]. Furthermore, it was shown that for many regimes of interest to ICF, the complex dynamical screening of the 2-component (electron + proton) plasma could be replaced by the static response of the electrons alone, yielding identical results [8, 9, 12, 13]. This is hardly surprising, since in a given electron-proton collision, the "spectator" electrons are expected to move much faster than the much heavier "spectator" protons. It is in this sense that a description of the collision rate in terms of a Coulomb-logarithm, $\ln \lambda_{ei} = \ln(b_{\max}/b_{\min})$, is sensible, where the maximum impact parameter, b_{\max} , is identified with the *static electron-only* Debye length. Such simplifications

in turn form the basis for analytic in-line models of electron-ion T -equilibration widely used in ICF simulation codes [1, 2].

However, as was pointed out early on by Dharma-wardana and Perrot [4, 17], this failure of the ions to participate in the screening may not hold in general. Indeed, coupled modes of the electron-ion system are known to exist in the form of ion acoustic waves (analogous to phonons in a solid), and their presence is expected to alter the equilibration rate relative to that predicted by a theory in which independent electron and ion plasma waves are solely responsible for the exchange of energy in a 2- T plasma. A regime in which coupled modes were argued to be important is that accessed in the intense laser irradiation of condensed matter [18]: solid density, and $T_e \gg T_i$. Roughly a decade after this work, Gregori, Gericke, Vorberger and coworkers (in various combinations) [19–21] used the Lenard-Balescu kinetic equation to study both the nature of the effect of coupled modes on T -equilibration, and the precise regime where they are expected to be important for weakly-coupled systems. By analyzing the detailed structure of the 2-component dynamical dielectric function, the authors of Ref.[20] presented this condition for the importance of coupled modes for a plasma with electrons and a single species of ions [22]:

$$T_i \lesssim 0.27 \cdot Z_i T_e, \quad (1)$$

for non-degenerate electrons; here, Z_i is the charge of the ion. For hydrogen ($Z_i = 1$), T_e must be at least 4 times larger than T_i for coupled modes to have an effect. This is not the typical scenario in ICF, at least within the region of undoped DT fuel. However, if Z_i is greater than 4, the above condition suggests that theoretical treatments which neglect coupled modes, such as Landau-Spitzer, may be suspect even for $T_e \sim T_i$.

In the work of Ref.[20], comparisons were made between the so-called coupled-mode theory (perturbative treatment of Lenard-Balescu including dynamical screening) and the Fermi Golden Rule (FGR) theory [4] (analogous treatment, but where the ion f-sum rule is used to reduce the screening to that of the static response of the electrons [8, 20]). The systems considered were hydrogen at a particle density, $n = 10^{26} \text{ cm}^{-3}$, and species temperatures: $T_i = 10^5 \text{ K}$, and $10^5 \text{ K} < T_e < 10^{10} \text{ K}$. It was predicted that the presence of CMs reduces the energy equilibration rate by up to $\sim 50\%$ relative to that predicted by the FGR theory and other approaches akin to Landau-Spitzer. The fact that it is a reduction is easy to understand: Ion-acoustic waves are modes in which a significant fraction of the electrons

form screening clouds which move with the ions. Such concerted motion is not conducive to resistive heating of ions by electrons (or vice versa), for precisely the same reason that energy transfer is not permitted in a simulation in which the Born-Oppenheimer approximation is invoked (electrons slaved to the ion dynamics). The work of Ref.[20] is the first we know of to attempt to fully quantify this effect for hydrogen plasmas.

As we have already implied, the presence of CMs renders the theoretical description sufficiently complex so that a simple analytic representation of the T -equilibration rate is no longer possible. The potential importance for ICF simulations, however, has prompted researchers to search for approximate expressions which capture the salient features of the reduction in the rate due to ion-acoustic waves [19, 23]. The work of Chapman et al. in Ref. [23] is particularly notable for its accuracy in this regard, but its application in ICF simulations [24] incurs significant computational expense relative to the simpler theories which neglect CMs such as Landau-Spitzer and its many variants [12, 13, 15].

Because the aforementioned predictions of the CM effect in T -equilibration used a variant of kinetic theory which is expected to be exact only in the limit of weak coupling [16], it is reasonable to ask if these predictions are accurate in practical realizations of out-of-equilibrium hydrogen plasmas. To this end, Vorberger and Gericke searched through the existing MD results [8, 9, 11–14, 25] to assess if simulations within the regime of interest for CMs had been performed [26]. Their conclusion was that only one simulation *might* have exhibited the CM effect [25]. This observation, together with the fact that experimental measurements of T -equilibration in plasmas are practically nonexistent, emphasizes the need for dedicated MD simulations specifically tailored to measure the effect of CMs on the relaxation rates. This would in turn aid in the development of more accurate in-line models for use in ICF codes, by buttressing our confidence in the theories used to construct such models [19, 23], and/or critiquing their assumptions if their predictions are shown to be inaccurate.

In this work, we study temperature equilibration with non-equilibrium classical MD simulations for plasmas which are chosen to lie squarely within the regime indicated by Eq.1: hydrogen at a density of $n = 10^{26} \text{ cm}^{-3}$ and $T_e \gg T_i$. These are chosen to coincide with the plasmas studied in Ref.[20]. We model the inter-particle interactions in these plasmas by Dunn-Broyles diffractive [27] and Minoo et al. exchange [28] QSPs [29]. For each plasma studied, we compare our MD results to both the coupled-mode Lenard-Balescu (CM-LB)

theory of T -equilibration and the FGR (neglecting CMs) theory. Since we invoke an approximate description of these hydrogen plasmas in which the electrons are classical particles that interact via effective T -dependent interactions, we take care to apply the CM-LB and FGR theories *to the classical QSP system* rather than to the quantum-Coulomb system as done in Ref.[20], to ensure that effects arising from the assumption of classical electrons and altered effective interactions are included appropriately [11].

We present and discuss several prominent MD results here, for these specific hydrogen cases:

1) The temperature relaxation is asymmetric, $dT_e/dt \neq -dT_i/dt$, indicating sizable contributions to dT_e/dt and dT_i/dt from time-varying potential energy, V . This effect is beyond the weak-coupling treatment of standard Lenard-Balescu (which only conserves kinetic energy), but it has been discussed and studied before in numerous theoretical contributions [3, 7, 21], and has been noted in at least one previous classical MD study [14]. We define and extract an effective "two-temperature thermodynamic" potential, $E(T_i, T_e)$ (with associated heat capacities, $C_e = \partial E / \partial T_e|_{T_i}$ and $C_i = \partial E / \partial T_i|_{T_e}$), from the MD, and show (for one of these hydrogen cases) that it can be understood from straightforward modeling. Additional predictions made with a quantum screened-Coulomb model and with Orbital-Free Density Functional theory indicate that these effects from time-varying potential energy should be noticeable for true quantum-Coulomb hydrogen plasmas in these conditions as well.

2) Despite this success, we see that the kinetic ion temperature [30] and the configurational ion temperature [31] disagree, in that the instantaneous static structure of the ions is *not* that as expected for plasmas with T_i as defined by the ion velocity distribution function. Rather, it is reminiscent of a plasma with a somewhat lower T_i . Nevertheless, the analysis in 1) succeeds because the derivatives, C_e and C_i , are relatively insensitive to the differences in temperatures; i.e., E is linear over wide ranges of T_e, T_i .

3) We determine the instantaneous ion velocity distribution in the MD during temperature relaxation, and quantify its departure from the equilibrium Maxwell-Boltzmann distribution. While this deviation does slightly affect the Lenard-Balescu prediction for temperature relaxation, explicit calculations using a polynomial expansion show that it should not significantly alter the equilibration rates (at least within the weak-coupling approximation inherent in Lenard-Balescu).

And most importantly,

4) We quantify the energy relaxation rate in MD by decomposing the total energy into species-dependent pieces, $E = E_i + E_e$, in which the electron-ion potential energy, V_{ei} , is shared equally between the species [21]. It is shown that dE_i/dt ($= -dE_e/dt$) is far closer to the FGR predictions than to the CM predictions, in stark contrast to expectations.

The remainder of this paper is organized as follows: Section II presents our MD methodology, including a discussion of the multiple methods we use to initialize our 2- T simulations. Section III contains a discussion of the perturbative treatment of the Lenard-Balescu equation used to predict the instantaneous time rates of change of electron and ion temperatures; here we focus on computational issues specific to predictions in the CM regime. A presentation of the results of our MD simulations and the comparison to the theoretical predictions (FGR and CM-LB) appears in Section IV; this includes a discussion of the sizable effects of time-dependent potential energy, a study of the (negligible) effects of non-Maxwellian velocity distributions, and a discussion of possible reasons for the greatly reduced effect of coupled modes as compared to predictions [20]. We conclude in Section V.

II. MOLECULAR DYNAMICS SIMULATIONS

Coupled mode temperature equilibration includes the possibility of non-degenerate electrons, making classical molecular dynamics (MD) simulations a useful comparator. We employ the ddcMD code [32] with the PPPM method [33] for large-scale charged-particle simulations [34]. We mostly consider a subset of hydrogen cases studied by Vorberger and Gericke [20], $\rho = 10^{26}/\text{cc}$, $T_i = 10^5\text{K}$, and $T_e = 10^7$ to $5 \times 10^8\text{K}$. The associated MD time steps range from 5×10^{-6} to 1×10^{-6} fs. Most simulations include 5.12×10^5 hydrogens, but the first case is studied using 4.096×10^6 hydrogens. This improves statistical sampling and reduces noise in the final results.

Even for non-degenerate electrons, the attractive electron-ion interaction requires some accommodation of quantum effects. Accordingly, we use the Dunn-Broyles diffractive quantum statistical potential (QSP) [27]. We include the two-body repulsive exchange piece introduced by Minoo et al. [28]. This prescription is used in the early temperature equilibration work of Hansen and McDonald [10] and much of our work on this subject [11, 13]. It is an extension of the potentials (designed to reproduce quantum equilibrium pair correlations using classical particles) to a non-equilibrium situation. The potentials are parametrized for

some desired target ion and electron temperatures, $T_i^{\text{targ}} \neq T_e^{\text{targ}}$. The results for species α and β are then:

$$V_{\alpha\beta}(r, T) = \frac{Z_\alpha Z_\beta e^2}{r} \left[1 - \exp\left(\frac{-2\pi r}{\Lambda_{\alpha\beta}}\right) \right] + k_B T \ln(2) \exp\left(\frac{-4\pi \ln(2) r^2}{\Lambda_{\alpha\beta}^2}\right) \delta_{\alpha e} \delta_{\beta e}, \quad (2)$$

where $\mu_{\alpha\beta} = m_\alpha m_\beta / (m_\alpha + m_\beta)$, and k_B is Boltzmann's constant. The QSP is dependent on temperatures through the de Broglie wavelength, $\Lambda_{\alpha\beta} = \sqrt{2\pi\hbar^2 / \mu_{\alpha\beta} k_B T^{QSP}}$. (The temperature, T^{QSP} , is taken to be T_i^{targ} when α and β are both ions, and simply T_e^{targ} if either is an electron.) In practice, these potential parameters are held constant at target values during a simulation even as the instantaneous temperatures vary widely with time. The time-independent Hamiltonian then ensures energy conservation for microcanonical simulations. Although this prescription is not in keeping with the behavior of true quantum electron wave-packets as the electrons are cooled or heated, our aim in this work is simply to evaluate and critique the use of various theories in predicting T -equilibration for systems with a *fixed* set of interaction potentials [35]. This was exactly the approach adopted in Ref.[11] as well.

Despite the well-defined Hamiltonian, a unique definition/preparation of a nonequilibrium two-temperature system may not always be feasible; specification of the effective temperatures leaves many degrees of freedom unconstrained. Practically when NVE (constant particle number, volume, and energy) microcanonical simulations are allowed to evolve freely for a sufficient time, systems of a particular total energy, E , can often be found that pass through a desired target state, $T_e^{\text{targ}} \neq T_i^{\text{targ}}$. This state can be considered unique as long as other out-of-equilibrium degrees of freedom set by the initial conditions have relaxed much faster than the global temperature difference. For example, intraspecies velocity equilibration can be much faster than electron-ion interspecies. This gives an effectively unique two-temperature state in weakly-coupled cases. However when the potential energy is substantial, differences between the configurational temperature [31] and the kinetic values [30] may exist and be relevant. In particular, the CM regime includes, by design, long wavelength, underdamped, quasi-harmonic ion acoustic waves (IAW). Being underdamped, these modes would also be only weakly coupled to anything that might serve as a thermal bath. Hence, it is reasonable to suspect that different wavelength IAW might evince different ef-

fective temperatures during equilibration, and that structure factors and radial distribution functions will not correspond to those of a unique temperature. Normally, this might be ignored on phase space arguments (for small $|k|$, the number of modes, $\propto |k|^3$, and the associated heat capacity is small), but the CM regime relies on having a sufficient number of slowly-equilibrating degrees of freedom that the overall interspecies equilibration is affected.

Signatures of the potential energy are broadly visible in our CM temperature relaxation simulations. If a system is not carefully prepared, there is a prominent underdamped oscillation in the ion kinetic temperature at the start of the NVE evolution. Similar effects are seen when the electron temperature is abruptly altered [36]; the resulting change in the screened ion-ion potential switches the asymptotic pair distribution to some new $g_{ii}(r)$. Transient evolution away from the old stationary ion distribution towards the new one includes underdamped oscillations from IAW. Such oscillations in our CM simulations are precisely compensated by variations in the ion potential energy, $V_i \equiv V_{ii} + V_{ie}/2$, clearly showing the role of IAW collective modes. (Similar transients are not seen in the electron subsystem, which has much weaker coupling.) Yet another signature of the potential energy is that the temperature relaxation rates are not equal and opposite, $N_i \frac{dT_i}{dt} \neq -N_e \frac{dT_e}{dt}$, even after transient oscillations are mitigated. This will be discussed in more detail in Sec. IV.

Because of these complications, we consider three distinct approaches to MD sample preparation. In a few test cases, we start from near-stationary states subject to separate, species-dependent thermostats, similar to past analyses. However in our hydrogen CM cases, the ions require exceptionally strong thermostats to keep them cold. A Langevin thermostat [37] has the advantage of driving each species towards a Maxwell-Boltzmann velocity distribution. Unfortunately, the strong Langevin drag term has a time constant much less than a typical IAW period, and so the IAW are severely overdamped. In equilibrium, a quasi-harmonic mode of wavenumber k should have an RMS-expected thermal amplitude given by equipartition of energy. But given the overdamped ion dynamics, transient density perturbations will persist for times that scale as $1/k^2$, meaning that long-wavelength density fluctuations do not fully equilibrate during a simulation. In contrast, the Berendsen thermostat [38] simply scales all particle velocities of a given species so that their total kinetic energy is instantaneously related to the thermostat, $K \equiv \frac{3}{2}k_B T$. In this case, quasiharmonic oscillations are largely undamped by the thermostat, and long-wavelength density fluctuations are left to equilibrate by means of weak, intrinsic anharmonicities. This will

give rise to different transient IAW behavior as the system relaxes. Thirdly, we prepare the most extreme two-temperature state possible so as to maximize the time for transient artifacts to decay. Systems are started with the ions in a simple cubic lattice, frozen in place at $T_i = 0$; i.e., all long wavelength IAW are initialized with exactly zero amplitude. The electrons are then equilibrated in a microcanonical MD simulation. Each initial state is characterized by its total energy, E , and is rigorously defined as part of a constrained, equilibrium ensemble. Once the ions are released, we expect monotonic and continuous temperature relaxation with time. In particular, the family of instantaneous equilibrating states can be completely characterized by any two of the variables, E, T_e, T_i . This is akin to having an effective, two-temperature thermodynamic potential, $E(T_e, T_i)$, which can then be used to compute derivatives by finite differencing across different microcanonical simulations. This last preparation method is used for the majority of the simulations because of this advantage.

For example, a hydrogen case with $n = 10^{26}/\text{cm}^3$, $T_p = 10^5$ K, and $T_e = 10^7$ K has been tested with all three preparation methods; all three give similar results. The total energy is tuned for each system until their electron temperatures are the same to within 0.1% when the target T_i is reached. The residual oscillations in $T_i(t)$ differ slightly in their magnitudes at the target conditions, but dT_i/dt can be measured across approximately one oscillation period to minimize any dependence on that amplitude. With that approach, the total potential energies at target are equal among the different preparation methods to within 0.1 eV/hydrogen (per hydrogen= per proton= per electron), and the measured dT_i/dt are 5.45, 5.45, and 5.48 eV/fs for lattice, Langevin, and Berendsen initial configurations, respectively. Thus any possible, long-lived differences in IAW amplitudes in the CM regime seem to have no effect on the potential energy of the system or the overall equilibration rates.

A typical result for the lattice preparation procedure is shown in Fig. 1 for the ion temperature and for a series of ion-ion pair correlation functions, $g_{ii}(r)$. At $t=0$, the constraint on the ions is released, and the ion temperature quickly begins to rise. Simultaneously, there are long-period, transient oscillations initially visible in T_i . The initial lattice structure is lost well before the target condition of $T_i = 8.6$ eV ($\sim 10^5$ K) is reached in Fig. 1. By that time, the remnants of the long-period oscillations are also only of order ± 5 meV. In practice, $T_i(t)$ at this time is closely linear plus a sinusoidal component; the slope at target is thus rendered insensitive to any long-period transient by making a linear fit over at least

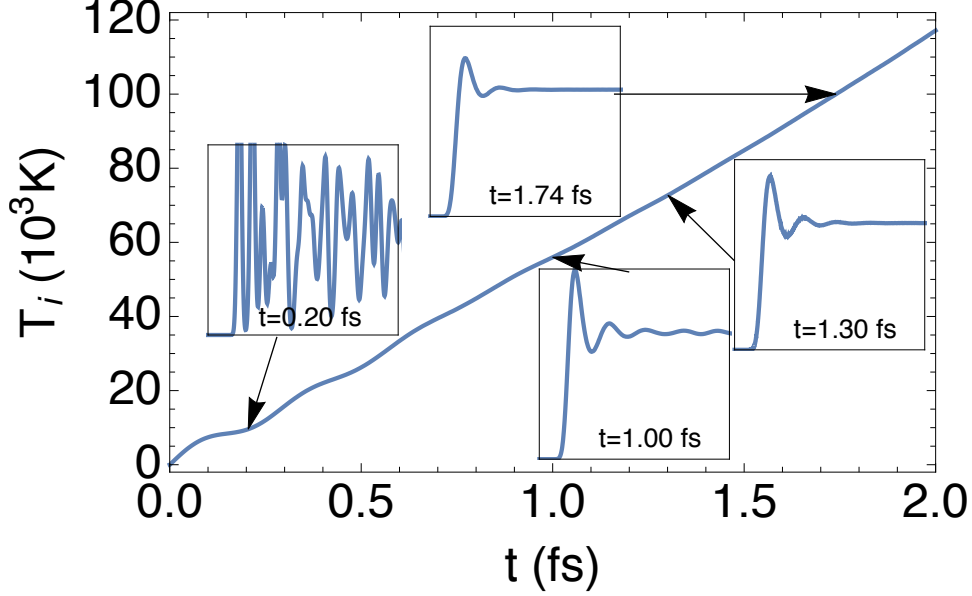


FIG. 1: Effective proton temperature, $T_i = 2/3\langle E_i^{kin} \rangle$, versus time for a single NVE run of 8.192×10^5 particles (protons plus electrons). Oscillations in T_i correspond to underdamped transient relaxations in the ion-ion radial distribution function. Insets show the pair distribution function, $g_{ii}(r, t)$; showing signs of crystalline order at early times. Long-range correlations are lost within 1.5 fs. The last inset shows the target conditions for this case, $T_p = 10^5$ K, $T_e = 10^7$ K, which occur at $t=1.74$ fs.

a complete cycle. Additionally, we note that the detailed shape of $g_{ii}(r)$ for times after the transient behavior has subsided is almost completely independent of the sample preparation scheme. In this sense, the non-adiabatic effects manifest in $g_{ii}(r)$ that we discuss later (in Section IV B) are also not the result of transient effects, and are therefore representative of uniquely-defined states.

For non-equilibrium plasmas, temperature relaxation is directly related to the underlying heat flow only in the weak-coupling limit. In the CM cases we study, $\Gamma_{ii} = 12.5$ ($= \frac{Z^2 e^2}{k_B T_i R}$ where $R = [\frac{3n}{4\pi}]^{1/3}$ is the average nearest-neighbor distance between ions), and the evolving potential energy is comparatively large. This evolution is associated with observed asymmetric temperature changes, $N_i k_B dT_i/dt \neq -N_e k_B dT_e/dt$. To better quantify this, we will expand the MD analysis to include estimates of $\left(\frac{\partial V}{\partial T_i}\right)_{T_e}$ and $\left(\frac{\partial V}{\partial T_e}\right)_{T_i}$, analogous to a two-temperature excess heat capacity.

A single NVE simulation can only be used to compute dV/dt (or i.e., the gradient along

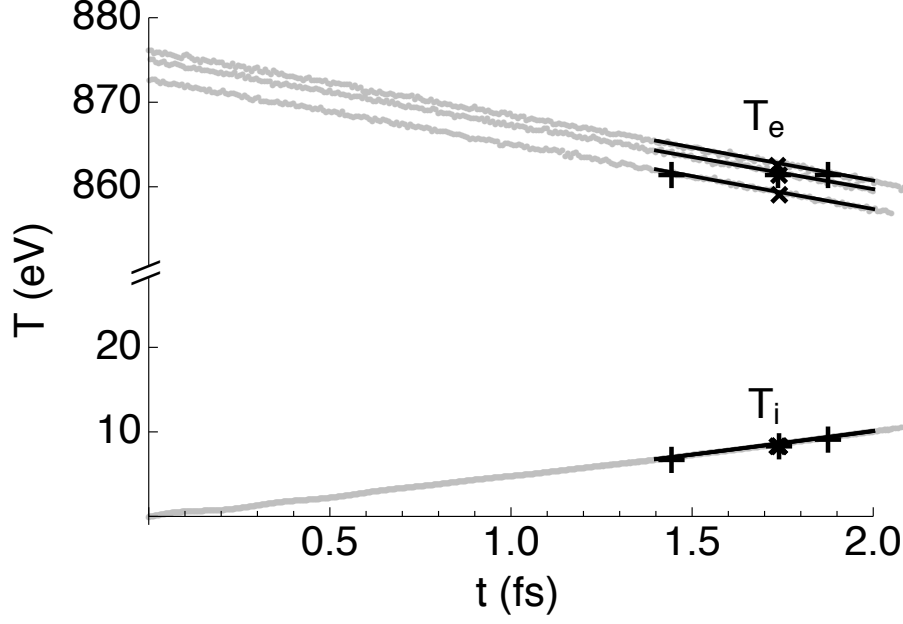


FIG. 2: Temperature histories are shown in light gray for three separate, microcanonical MD simulations differing in total energy by a few eV/e-i pair. The temperature curves appear noisy because of intrinsic, short-time fluctuations that are easily averaged over. Linear least squares fits to portions of the time series are shown as black lines; dT/dt is computed analytically from these fits. The fitted curves each reach the target ion temperature $T_p = 10^5$ K) at times marked by \times . They reach the target electron temperature ($T_e = 10^7$ K) at times marked with $+$. By design, these times coincide for the middle microcanonical system.

the prescribed temperature trajectory, $\vec{\nabla}V \cdot (dT_i/dt, dT_e/dt)^\top$. Other derivatives in T_e , T_i space require cross-comparison of different microcanonical ensembles. Fig. 2 shows the temperature histories of three separate, microcanonical simulations, spanning a range in E of about 4 eV/hydrogen. Each system is prepared with a lattice of ions at zero temperature; given the different total energies, the initial electron temperatures are well-separated among the three systems. The middle T_e curve corresponds to the unique system total energy for which T_e and T_i both reach their respective target values at essentially equal times. The adjacent curves approximately cover some range $T_e(t=0) - \Delta$ to $T_e(t=0) + \Delta$. Since the change to each system is small, $\Delta \ll T_e - T_i$, the relaxation rates, dT_e/dt , are nearly equal for the three runs, and the curves are nearly parallel. Similarly, because the $T_i(t=0)$ initially coincide, all three ion temperature curves are nearly indistinguishable.

As discussed above, a linear, least-squares fit is made to each temperature series, $T(t)$,

to suppress any transient oscillations; this is shown in Fig. 2 with short segments in black. We analytically obtain dT/dt at the target conditions from these fits. The \times and $+$ symbols in Fig. 2 also mark the times along each fitted curve when the target temperature value is reached for ions or electrons, respectively. The instantaneous states at the marked points can then be used to compute temperature-dependent derivatives. Using the \times points, we would obtain $E(T_i = 10^5 \text{ K}, T_e)$ at three distinct T_e , while the $+$ points give $E(T_i, T_e = 10^7 \text{ K})$ at three values of T_i . In practice, $E(T_i, T_e)$ is linear in T_e and T_i (and vice versa) for a family of instantaneous states over a small range of energies, $[E - \Delta : E + \Delta]$, for Δ of order 1 eV per hydrogen. Thus, we apply another linear least squares fit to $E(T_i)|_{T_e=10^7}$ or $E(T_e)|_{T_i=10^5}$ for multiple microcanonical systems within ± 1 eV/hydrogen. We then compute $\partial E/\partial T$ analytically. Since the kinetic energy component satisfies $K \equiv \frac{3}{2}k_B T$, the resulting derivatives also give estimates for $\left(\frac{\partial V}{\partial T_i}\right)_{T_e}$ and $\left(\frac{\partial V}{\partial T_e}\right)_{T_i}$.

III. LENARD-BALESCU THEORY

To analyze our MD results, as well as to exercise and test existing theories of the CM effect in T -equilibration, we employ the Generalized Lenard-Balescu (GLB) theory [16] outlined in detail in Ref.[8]. In this approach, the time rate of change of the ion temperature, dT_i/dt , in a 2- T plasma is related via the fluctuation-dissipation theorem to the density response functions of the individual species. For the case of two species, electrons and ions, the result is [8]:

$$\begin{aligned} \frac{dT_i}{dt} = & -\frac{\hbar}{3\pi^3 n_\alpha} \int_0^\infty k^2 dk \int_0^\infty \omega d\omega \left[\frac{v_{ei}(k)}{D(k, \omega)} \right]^2 [1 - G_{ei}(k, \omega)] \times \\ & \left[N \left(\frac{\hbar\omega}{2k_B T_i} \right) - N \left(\frac{\hbar\omega}{2k_B T_e} \right) \right] \text{Im}\chi_e^0(k, \omega) \text{Im}\chi_i^0(k, \omega), \end{aligned} \quad (3)$$

where $D(k, \omega)$ is the 2-species dielectric function,

$$\begin{aligned} D = & [1 - v_{ee}(1 - G_{ee})\chi_e^0][1 - v_{ii}(1 - G_{ii})\chi_i^0] \\ & - v_{ei}^2(1 - G_{ei})(1 - G_{ie})\chi_e^0\chi_i^0, \end{aligned} \quad (4)$$

involving the inter-particle interactions v , free particle density response functions χ^0 , and Local Field Corrections (LFCs) G . $N(x)$ is a statistical factor arising from the fluctuation-dissipation theorem, and is equal to $\coth(x)$ for Fermions and $1/x$ for classical particles [39].

The response functions and LFCs are defined by the equation,

$$\delta n_\alpha = \delta n_\alpha^0 + \chi_\alpha^0 \sum_\beta v_{\alpha\beta} [1 - G_{\alpha\beta}] \delta n_\beta, \quad (5)$$

where the argument of each quantity is (k, ω) . Here, δn_α^0 are the density fluctuations in the absence of interactions, while δn_α are the density fluctuations in the presence of interactions [8, 39], with species indexed by α and β .

Eq.5 expresses the self-consistent rearrangement of charge resulting from screening in the multi-component plasma; if the LFCs ($G_{\alpha\beta}$) are set to zero, this is the Random Phase Approximation (RPA). The treatment of Eq.3 with LFCs set to zero is equivalent to the Lenard-Balescu theory of T -equilibration used in Refs.[7, 20, 21, 23]. This is the theory we use to compare to MD for our hydrogen cases where QSPs have been used. For the like-charge Coulomb cases, it is essential for us to use non-zero LFCs (particularly G_{ei}) in order for the integral over k in Eq.3 to be finite for large- k [8]. These implementations of GLB are identical to those we made in an earlier work [11].

The free-particle response functions, χ^0 , appearing in Eqs.3, 4, and 5 are the usual ones [39],

$$\chi_\alpha^0(k, \omega) = \lim_{\eta \rightarrow 0^+} \left[2 \sum_{\mathbf{k}'} \frac{f_\alpha(\mathbf{k}' + \mathbf{k}) - f_\alpha(\mathbf{k}')}{\frac{\hbar^2(\mathbf{k}'+\mathbf{k})^2}{2m_\alpha} - \frac{\hbar^2 k'^2}{2m_\alpha} - \hbar\omega - i\eta} \right], \quad (6)$$

where $f_\alpha(\mathbf{k})$ is the occupation number for particles of energy $\hbar^2 k^2 / 2m_\alpha$ at the temperature T_α . For our practical implementation for quantum particles, we use the Pade approximant fits for χ^0 appearing in Dandrea, Ashcroft, and Carlsson [40]; for classical particles, we use the analytic expressions found in Ref.[41]. Since we have shown in the past [11] that the inclusion of (static, in that case) LFCs does not improve agreement with MD for T -equilibration rates for hydrogen plasmas as modeled with QSPs, we set the $G_{\alpha\beta} = 0$ in the remainder of this work.

As discussed in numerous works (e.g., Refs.[7, 8, 11]), the expression of Eq.3 reduces to that of far simpler expressions in many situations of interest. First, if the ions are taken to be much more massive than the electrons, as is physically the case, *and* the conditions are *not* those described by Eq.1 [20], the f-sum rule for ions can be used to perform the ω -integral analytically by evaluating the electron quantities at $\omega \approx 0$. This is strictly correct when CMs are neglected, and arises from the fact that the plasma frequency of electrons far exceeds that of the ions, leading to the FGR result [4, 20]. This FGR expression, now

a single integral over k (see for instance Eq.12 in Ref.[8]), can then be massaged into the familiar Landau-Spitzer equilibration rate in the limit of weak plasma coupling [8]:

$$\frac{dT_i}{dt} = \frac{T_e - T_i}{\tau_{ei}}, \quad (7)$$

where

$$\frac{1}{\tau_{ei}} = \frac{8\sqrt{2\pi}n_i Z_i^2 e^4}{3m_e m_i} \left[\frac{k_B T_e}{m_e} + \frac{k_B T_i}{m_i} \right]^{-3/2} \ln \lambda_{ei}, \quad (8)$$

and $\ln \lambda_{ei} = \ln(b_{\max}/b_{\min})$, as described above. Here, the screening length, b_{\max} , arises from the small- k part of the dielectric function, D , in Eq.3. The b_{\min} comes from either the large- k part of $\text{Im}\chi_e^0$ (de Broglie wave length; quantum-Coulomb case), or the large- k part of $1 - G_{ei}$ (classical turning-point; classical-Coulomb case) [8]. For the classical-QSP case, b_{\min} arises from the large- k behavior of $v_{ei}(k)$ (\sim de Broglie wave length) [11]. The expressions of Eqs.7 and 8, as well as the full GLB expression of Eq.3, respect *kinetic* energy conservation, consistent with the assumption of weak plasma coupling.

If CMs are important, as is predicted to be the case for 2-component plasmas satisfying Eq.1, a reduction of Eq.3 to a Landau-Spitzer-like expression is not possible [23]. However, as long as the plasma is not too strongly-coupled, it is expected that the full double-integral expression of Eq.3 should still produce reliable predictions, and indeed it is essentially this theory which was used by Vorberger and Gericke to derive the regime indicated by Eq.1. In Ref.[20], these authors emphasized that the correct inclusion of CMs (in the form of ion acoustic waves) in T -equilibration within Lenard-Balescu is quite numerically challenging, because the structure of the ω -integrand possesses peaks which are infinitesimally sharp at small- k , where the effect is most important. This can be seen most easily by examining the structure of the energy loss function, $\mathcal{L}(k, \omega) \equiv \text{Im}[1/D(k, \omega)] = -\text{Im}[D]/([\text{Im}D]^2 + [\text{Re}D]^2)$. If v_{ei} is set to zero, this function has separate electron and ion plasmon peaks at the respective plasma frequencies, when $\text{Re}D = 0$ and $\text{Im}D$ is very small. However when $v_{ei} \neq 0$, the low- k portion of the ion plasmon sharpens while losing its intensity, and moves to much lower frequency, ceasing to contribute substantively to the dT_i/dt integral.

We illustrate the contributions to this integral by examining the k -integrand, $F(k)$, defined such that

$$\frac{dT_i}{dt} = \int_0^\infty F(k) dk, \quad (9)$$

i.e., the k -dependent function that results from performing the ω -integral in Eq.3. Figure 3 shows $F(k)$ computed for the (quantum-Coulomb) hydrogen plasma (here, taking all $G_{\alpha\beta}=0$) with $n = 1.0 \times 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, and $T_e = 10^7 \text{ K}$. The blue curve is the result of the FGR theory, which assumes no CMs and arises from Eq.12 in Ref.[8], reproduced here as:

$$\frac{dT_i}{dt} = \frac{(T_i - T_e)}{3\pi^2 m_i} \int_0^\infty dk k^4 \left[\frac{v_{ei}(k)}{\epsilon_e(k, 0)} \right]^2 \left(\frac{\partial \text{Im} \chi_e^0}{\partial \omega} \right)_{\omega=0}, \quad (10)$$

where ϵ_e is the one-component RPA plasma dielectric function of the electrons ($\epsilon_e = 1 - v_{ee}\chi_e^0$). The red curve shows $F(k)$ computed with Eq.3 using the numerical prescription outlined in Ref.[11], in which a logarithmic ω -mesh is used to compute the ω -integral, but no special attention is paid to ensuring that the ω -poles for small k are resolved properly [42]. The sharp peaks and valleys in $F(k)$ at the smaller k values arise from the improper treatment of the pole for which D is very close to zero; as k is changed, the pole alternately moves close to and farther from the nearest ω -mesh point, resulting in inaccurate integration. This can be corrected by treating the pole explicitly [20]. In our simple treatment here, we approximate this feature as a Lorentzian peak in the loss function, $L(x) = \frac{1}{\pi} \frac{\Gamma/2}{(x-x_0)^2 + (\Gamma/2)^2}$. While $\mathcal{L}(k, \omega)$ doesn't appear directly in Eq.3, multiplying and dividing by $\text{Im}D$ allows us to rewrite Eq.3 in the following way:

$$\begin{aligned} \frac{dT_i}{dt} &\propto \\ &\int_0^\infty v_{ei}^2(k) k^2 dk \int_0^\infty \frac{\omega d\omega}{|D(k, \omega)|^2} \Delta N(\omega) \text{Im} \chi_e^0(k, \omega) \text{Im} \chi_i^0(k, \omega) \\ &\equiv \int_0^\infty v_{ei}^2(k) k^2 dk \cdot I(k), \end{aligned}$$

where

$$\begin{aligned} I(k) &= \int_0^\infty \frac{\omega d\omega \Delta N(\omega) \text{Im} \epsilon_e \text{Im} \epsilon_i}{|D(k, \omega)|^2} \\ &= \int_0^\infty \frac{\omega d\omega \Delta N(\omega) \text{Im} \epsilon_e \text{Im} \epsilon_i}{\text{Im} D} \cdot \left[\frac{\text{Im} D}{|D(k, \omega)|^2} \right] \\ &\equiv \int_0^\infty d\omega R(k, \omega) \cdot \mathcal{L}(k, \omega). \end{aligned}$$

Here, ΔN is the difference of statistical factors appearing in Eq.3, and the individual species dielectric functions are defined by,

$$\epsilon_e = 1 - v_{ee}\chi_e^0,$$

$$\epsilon_i = 1 - v_{ii}\chi_i^0.$$

$R(k, \omega)$ is the ω -integrand of Eq.3, but with the loss function factored out. $\mathcal{L}(k, \omega) \rightarrow L(\omega - \omega_0) \sim \delta(\omega - \omega_0)$ when $\text{Re}[D]$ goes through 0 and $\text{Im}[D] \rightarrow 0$. Thus,

$$I(k) \rightarrow \frac{\pi}{A} R(k, \omega_0),$$

when $\text{Re}[D] \sim A(\omega - \omega_0)$. The width of the Lorentzian is $\text{Im}D(\omega_0)/A$, and can be compared to the quadrature mesh size, $\Delta\omega$. We then take the corrected integral to be: [Uncorrected integral on the ω -mesh] - [integral of the approximated form on the ω -mesh] + [analytically-derived correction]. This strategy produces the green curve in Fig.3. Note that this curve still possesses peaks and valleys at the identical locations to those of the red curve, but their magnitudes are greatly suppressed. The remaining bumps are an artifact of the approximation, $\mathcal{L}(k, \omega) \rightarrow L(\omega - \omega_0)$; a better representation of the true *asymmetrically* broadened ion acoustic wave peak yields still smoother results [20, 23]. However, we note that the final integral, $\int_0^\infty F(k)dk$, is *equal to within 0.1%* for red and green curves. Thus, the main feature of CMs, the extinguishing of the small- k contributions to dT_i/dt (relative to that of the FGR theory; see Fig.3) [20], is captured even for a naive evaluation of Eq.3. This is less true as T_e is raised and the ratio T_e/T_i is increased; we have noted up to 10% deviations in some cases, when using the naive evaluation. Therefore all of the results we present here make use of the pole-correction strategy above; we will see below that they agree quantitatively with the predictions of Ref.[20] for quantum-Coulomb hydrogen plasmas.

Since we are (a) reinvestigating prior predictions for quantum plasmas, and are (b) comparing to the results of our classical MD studies with QSPs, we carry forth the above GLB prescription in two modes: 1. with quantum statistics and quantum response functions together with the bare Coulomb interaction, and, 2. with classical statistics and classical response functions together with the QSPs. This is in the spirit of our earlier MD/theory work on T -equilibration *not* in the CM regime [11], where we found that the use of Dunn-Broyles QSPs and classical statistics substantively reduced the equilibration rates relative to the quantum-Coulomb predictions [43]. Because our aim in this work is to use MD to validate theories which have predicted the CM effect in T -equilibration, it is crucial for us to separate possible reductions in rates resulting from the use of classical MD and QSPs from the actual coupled-mode effect itself. In this sense, the classical-QSP system is to be thought of as a theoretical surrogate in which the CM effect is studied. If agreement between classical-QSP MD and classical-QSP Lenard-Balescu is demonstrated, our confidence in the

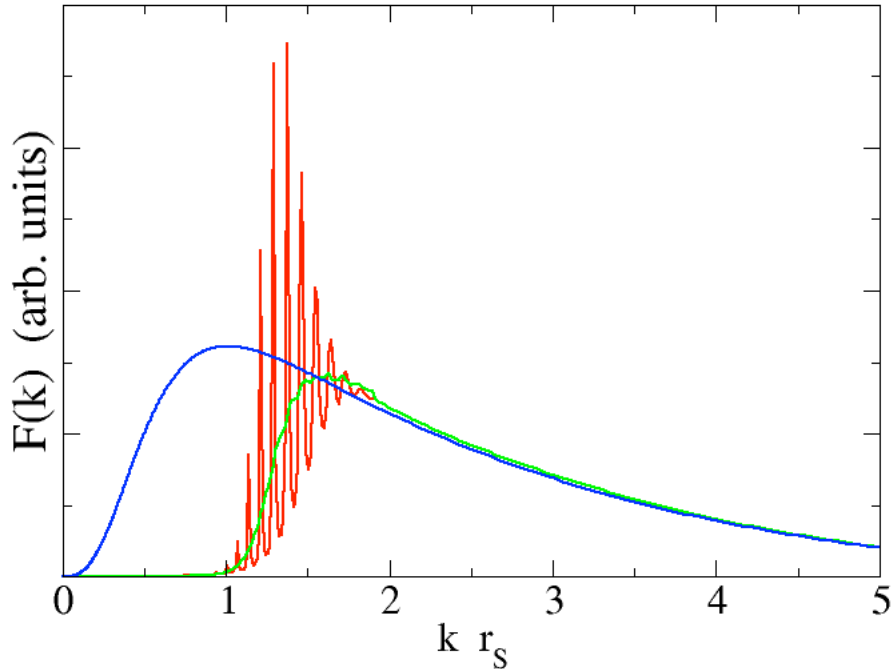


FIG. 3: The k -integrands, $F(k)$, of various Lenard-Balescu expressions for the equilibration rate for the quantum hydrogen plasma with $n = 1.0 \times 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, and $T_e = 10^7 \text{ K}$. Red curve: $F(k)$ computed by Eq.3, using the "naive" evaluation scheme outlined in Ref.[11]. Green curve: $F(k)$ from Eq.3, but computed with the pole-correction scheme outlined here. Blue curve: $F(k)$ from the FGR expression of Eq.10. The integrals under the two coupled-mode (CM) Lenard-Balescu $F(k)$ curves shown here (red, green) are equal to within 1% for this case.

quantum Lenard-Balescu predictions of the CM effect for real plasmas is strengthened.

IV. RESULTS AND DISCUSSIONS

A. MD results for the Vorberger-Gericke [20] hydrogen plasmas ($n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, $10^7 \text{ K} < T_e < 10^9 \text{ K}$)

We report T -equilibration for the family of hydrogen plasmas with density $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, and T_e between 10^6 K and $5 \times 10^8 \text{ K}$. These plasmas were considered in Ref.[20] using LB, and were predicted to equilibrate at rates substantially slower than those

T_e (eV)	1×10^7	2×10^7	3×10^7	5×10^7	1×10^8	2×10^8	5×10^8
MD ion	5.63 ± 0.05	6.65 ± 0.03	6.92 ± 0.06	6.77 ± 0.02	6.09 ± 0.04	5.42 ± 0.02	4.32
MD ele	7.78 ± 0.06	9.72 ± 0.08	10.30 ± 0.08	10.37 ± 0.05	9.63 ± 0.11	8.49 ± 0.09	6.75
MD E	12.46 ± 0.04	15.43 ± 0.05	16.25 ± 0.03	16.28 ± 0.04	14.83 ± 0.11	13.10 ± 0.07	10.45 ± 0.08
LB-CM	4.839	6.027	6.321	6.305	5.818		
LB-FGR	9.840	11.475	11.732	11.474	10.451		

TABLE I: Temperature relaxation rates, $|dT/dt|$. All units are eV/fs. $E_i = 3/2kT_i + V_i$ where $V_i = V_{ei}/2 + V_{ii}$, and $|dE_i/dt| = |dE_e/dt|$. When the energy rate of change is plotted in Fig. 5, it is expressed as $\frac{2}{3}|dE/dt|$ to compare to temperature derivatives.

T_e (eV)	1×10^7	2×10^7	3×10^7	5×10^7	1×10^8	2×10^8	5×10^8
$dV/dT_p _{T_e}$	0.629 ± 0.03	0.698 ± 0.03	0.765 ± 0.05	0.810 ± 0.04	0.895 ± 0.04	0.915 ± 0.04	0.832 ± 0.06
$dV/dT_e _{T_p}$	0.027 ± 0.02	-0.027 ± 0.02	0.00 ± 0.03	0.00 ± 0.03	0.016 ± 0.02	0.12 ± 0.08	-0.026 ± 0.03

TABLE II: Excess heat capacity in eV/fs. Species temperature is defined by $3/2kT \equiv$ kinetic energy; the ideal heat capacity is $3/2$.

as predicted by the FGR approach and a variant of Landau-Spitzer.

First we discuss our version of the LB predictions for these cases. The solid curves of Figure 4 display the energy transfer rate per electron, as a function of T_e , computed in two ways: 1. Solid red line: with the CM Lenard-Balescu theory (Eq.3 with zero LFCs), and 2. Solid blue line: with the FGR theory (Eq.10). Both curves result from the quantum-Coulomb evaluation of these equations. These curves compare very favorably with the corresponding curves in Fig.4 of Ref.[20]. The only notable discrepancy is at low- T_e , and is due to the fact that our FGR evaluation neglects electron degeneracy, unlike that of Ref.[20]. As indicated in the previous Section, our numerical treatment of the sharp low- k poles in the dielectric function of Eq.3 is sufficiently accurate to agree with the more carefully resolved results of Ref.[20]. The dot-dashed curves of Figure 4 show our calculations for the same plasmas, but for the *classical-QSP* cases, assuming Dunn-Broyles diffractive terms [27] and Minoo et al. exchange contributions [28] evaluated at the electron temperatures, T_e . These are the QSPs used for our classical MD studies for these plasmas, to be discussed directly below.

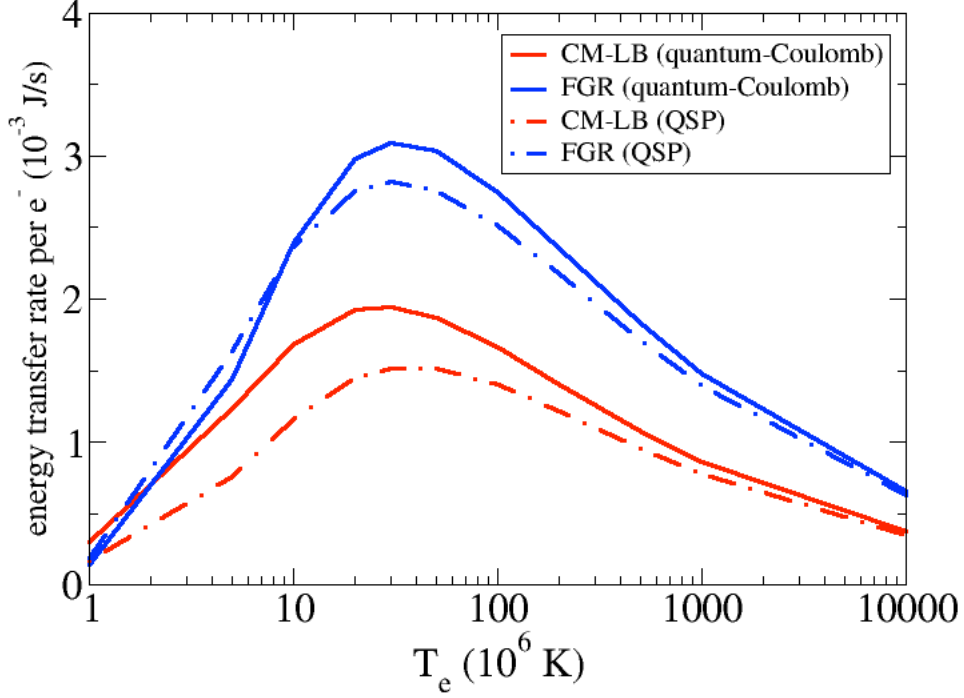


FIG. 4: Instantaneous energy transfer rate per e^- for hydrogen plasmas with $n = 10^{26} \text{ cm}^{-3}$ and $T_i = 10^5 \text{ K}$, as a function of T_e . Solid red curve: quantum-Coulomb prediction using the CM Lenard-Balescu approach; solid blue curve: quantum-Coulomb prediction with the FGR prescription. Dot-dashed red curve: classical-QSP prediction using the CM Lenard-Balescu approach; dot-dashed blue curve: classical-QSP prediction with the FGR prescription.

The red dot-dashed curve is the CM-LB result, while the blue dot-dashed curve is the FGR prediction. Note that for both curves, the energy transfer rates are substantially lower than those of the quantum-Coulomb results plotted in Fig.4. This reduction is very similar to that noted for the hydrogen cases studied in Ref.[11] when comparing quantum-Coulomb to classical-QSP Lenard-Balescu predictions (for the same choices of QSPs).

Fig.5 shows the predictions of Fig.4 once again, but now expressed as $|dT/dt|$ for the species rather than as an energy transfer rate. Also displayed are our MD results obtained using the ion-lattice preparation method discussed in Section II. We extract these rates as in Fig. 2, by fitting the MD time series around the target conditions; they are therefore some effective time-average of noisy MD dT_i/dt , $-dT_e/dt$, and dE/dt , respectively. We

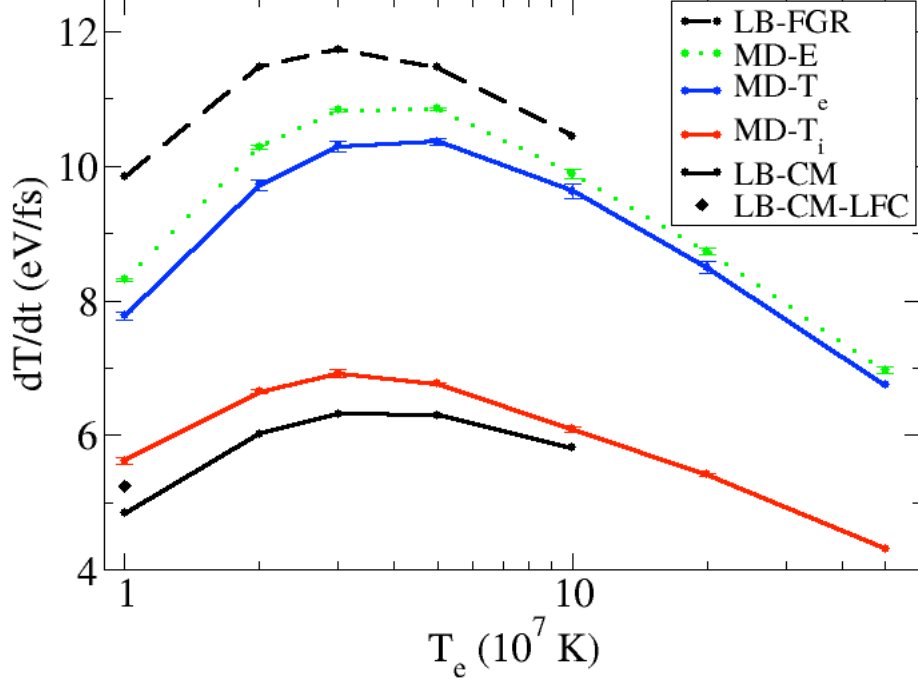


FIG. 5: Instantaneous dT/dt calculated from classical-QSP Lenard-Balescu, and extracted from MD using the identical QSPs for hydrogen plasmas with $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, and T_e as shown on the x-axis. The uppermost, thick, dashed, black curve is for LB-FGR, and the lowest, thick, solid, black curve is for LB-CM theory. The disconnected black point corresponds to a local field corrected LB-CM calculation using two-temperature HNC. Only the ion-ion local field correction is included. Red symbols with error bars and thin solid lines correspond to MD for dT_i/dt , and blue symbols with error bars and thin dashed lines correspond to MD for $-dT_e/dt$. The green symbols with error bars and thin dotted lines show the MD results for the time rate of change of total energy, scaled by $2/3$ from Table IV A to correspond to a "temperature", i.e. $\frac{2}{3}dE_e/dt$. (The relation $dE_i/dt = -dE_e/dt$ holds exactly).

note that dT_i/dt from the MD is slightly higher than the prediction from Lenard-Balescu including CMs, while $-dT_e/dt$ is far higher still. The two MD temperature derivatives approach equality for more weakly-coupled plasmas; indeed, essentially symmetric relaxations ($dT_e/dt = -dT_i/dt$) were noted in earlier work on hydrogen plasmas [8, 9, 11–13]. In our cases here, however, the ions are sufficiently cold that such an asymmetric temperature relax-

ation is expected [7, 21, 23]. This results from the sizable potential energy in the (somewhat strongly-correlated) screened ions, which diminishes as the ions are heated. The effect on dT_e/dt and dT_i/dt is simple to analyze using conservation of energy, within the rubric of the 2- T hypothesis we have invoked.

Since, after initial preparation, our MD simulations represent closed systems which evolve microcanonically, the total system energy, E , is conserved, and we have:

$$E(T_e, T_i) = K_e(T_e) + K_i(T_i) + V(T_e, T_i), \quad (11)$$

$$\frac{dE}{dt} = 0$$

$$= \left[\frac{3}{2}N_e + \left(\frac{\partial V}{\partial T_e} \right)_{T_i} \right] \frac{dT_e}{dt} + \left[\frac{3}{2}N_i + \left(\frac{\partial V}{\partial T_i} \right)_{T_e} \right] \frac{dT_i}{dt}, \quad (12)$$

where K_e and K_i are electron and ion kinetic energies, and are functions only of their respective temperatures, and V is the total system potential energy, which we assume to be a unique function of T_e and T_i [44]. Equating the two summands gives,

$$\left[\frac{3}{2}N_e + \left(\frac{\partial V}{\partial T_e} \right)_{T_i} \right] \frac{dT_e}{dt} = - \left[\frac{3}{2}N_i + \left(\frac{\partial V}{\partial T_i} \right)_{T_e} \right] \frac{dT_i}{dt}, \quad (13)$$

which leads to a prediction for the asymmetry in the T -derivatives:

$$\frac{\left| \frac{dT_e}{dt} \right|}{\left| \frac{dT_i}{dt} \right|} = \frac{\left[\frac{3}{2}N_i + \left(\frac{\partial V}{\partial T_i} \right)_{T_e} \right]}{\left[\frac{3}{2}N_e + \left(\frac{\partial V}{\partial T_e} \right)_{T_i} \right]}. \quad (14)$$

For our hydrogen cases, $N_e = N_i$, and since Lenard-Balescu conserves only *kinetic energy*, our LB prediction (black curve in Fig.5) is consistent with the $V = 0$ version of Eq.14,

$$\frac{\left| \frac{dT_e}{dt} \right|_{\text{LB}}}{\left| \frac{dT_i}{dt} \right|_{\text{LB}}} = \frac{N_i}{N_e} = 1. \quad (15)$$

The asymmetry in the T -relaxation is due to the nonzero V , and in particular to the specific derivatives of V which appear in Eq.14.

Before we discuss these potential energy derivatives further, we note that our MD results for *energy relaxation*, $dE_i/dt = -dE_e/dt$, are far closer to the FGR predictions than to those of Lenard-Balescu when coupled-modes are included. This is seen in Fig.5, in which

the dotted green line and circles display the MD values for $2/3 \times dE_i/dt$ (with $k_B = 1$), where the species-specific internal energies are defined by Eq. 16 exactly as in Ref.[21]:

$$E_i = K_i + V_{ii} + \frac{1}{2}V_{ei},$$

$$E_e = K_e + V_{ee} + \frac{1}{2}V_{ei}. \quad (16)$$

While the LB predictions (either CM or FGR) are only consistent with $E_i = K_i$ and $E_e = K_e$, it is reasonable to expect that the energy transfer rates might be well predicted by LB, since the primary drivers for the energy exchange, Coulomb collisions, are included therein. Indeed, it is for this reason that the LB predictions of Ref.[20] were represented as energy transfer (rather than temperature equilibration) rates; those authors left open the possibility that non-ideal specific heats would have to be included to relate the $dE_{e,i}/dt$ from LB to the $dT_{e,i}/dt$, as they did subsequently in their work of Ref.[21]. However, our MD results for these cases clearly show the $dE_{e,i}/dt$ themselves to be in disagreement with the coupled-mode LB predictions. This is discussed further in Section IV.C. We emphasize that these conclusions can only be drawn by making the proper comparisons: classical-QSP LB vs. classical-QSP MD [11].

B. Elucidation of 2-T equation of state for the $T_e = 10^7$ K case

In this subsection, we explore one of the particular cases studied above: $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5$ K, and $T_e = 10^7$ K; this system has intra-species plasma coupling constants $\Gamma_{ee} \sim 0.125$, and $\Gamma_{ii} \sim 12.5$. To establish that the large discrepancies between dT_i/dt and $-dT_e/dt$ in the MD results, as shown in Fig.5, can be accounted for by considering the $\left(\frac{\partial V}{\partial T_i}\right)_{T_e}$ and $\left(\frac{\partial V}{\partial T_e}\right)_{T_i}$ in Eq.14, we extract these quantities independently using two approaches: 1. from additional molecular dynamics simulations, and 2. from (adiabatic, quasistatic) theoretical estimates. Regarding 1, potential energy derivatives at *fixed* T_e or T_i can be inferred from families of T -equilibration MD simulations performed at closely-spaced initial conditions, as described in Sec. II. This produces the values $\left(\frac{\partial V}{\partial T_i}\right)_{T_e} = 0.586k_B/e^-$ and $\left(\frac{\partial V}{\partial T_e}\right)_{T_i} = 0.0328k_B/e^-$ for this case. Our values for $|dT_e/dt|$ and dT_i/dt are 7.92 eV/fs and 5.59 eV/fs respectively, giving a LHS of Eq.14 of 1.42; plugging our values for $\left(\frac{\partial V}{\partial T_i}\right)_{T_e}$ and $\left(\frac{\partial V}{\partial T_e}\right)_{T_i}$ into the RHS gives 1.36.

Regarding the second (theoretical) approach, V can be computed from the three radial distribution functions, $g_{ee}(r)$, $g_{ii}(r)$, and $g_{ei}(r)$, since the inter-particle QSPs are 2-body in nature [45]:

$$V = \frac{n_{\text{tot}}}{2} \sum_{\alpha, \beta} x_{\alpha} x_{\beta} \int d^3r g_{\alpha\beta}(r) v_{\alpha\beta}(r), \quad (17)$$

where α and β indicate species (e, i), $n_{\text{tot}} = \sum_{\alpha} n_{\alpha}$, and $x_{\alpha} = n_{\alpha}/n_{\text{tot}}$. We use a 2-temperature variant [46] of the Hypernetted Chain (HNC) approximation [47] to determine the $g_{\alpha\beta}(r)$ to be input into this expression. Here, the equations have been generalized to include mass-weighted temperatures $T_{ij} = (m_j T_i + m_i T_j)/(m_i + m_j)$, which is roughly equal to T_e for this case. This approximation is valid for systems in which each species can be described by distinct Maxwellians [48], and both the mass discrepancies ($m_e \ll m_i$) and temperature discrepancies ($T_e \gg T_i$) of the systems of interest in our work reinforce this assumption. With these HNC-derived $g_{\alpha\beta}(r)$ and Eq.17 [49], V is determined at the nominal $T_i = 10^5$ K and $T_e = 10^7$ K. Repeating the whole calculation at $T_i + \delta T_i$ and $T_e + \delta T_e$ then produces $\left(\frac{\partial V}{\partial T_i}\right)_{T_e} = 0.5352 k_B/e^-$ and $\left(\frac{\partial V}{\partial T_e}\right)_{T_i} = 0.009728 k_B/e^-$. While $\left(\frac{\partial V}{\partial T_e}\right)_{T_i}$ is rather different from that of the direct MD determination outlined above, $\left(\frac{\partial V}{\partial T_i}\right)_{T_e}$ is quite similar; the smallness of $\left(\frac{\partial V}{\partial T_e}\right)_{T_i}$ relative to $3/2$ then results in the prediction $\left|\frac{dT_e}{dt}\right| / \left|\frac{dT_i}{dt}\right| = 1.33$, once again slightly below the MD value of 1.42.

While the aforementioned theory-MD agreement is reasonable for $\left(\frac{\partial V}{\partial T_i}\right)_{T_e}$ and $\left(\frac{\partial V}{\partial T_e}\right)_{T_i}$, it is interesting to look directly at the $g_{\alpha\beta}(r)$ from the MD, since our HNC calculations of these quantities are necessarily somewhat approximate. Fig.6 shows g_{ee} , g_{ei} , and g_{ii} for the $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5$ K, $T_e = 10^7$ K case. Solid lines show the results of our QSP-MD, where the average over snapshots was performed over a duration small enough to render T_e and T_i determinations essentially instantaneous. The dashed lines show our HNC calculations for this case using the identical QSPs; these are the functions which we input into Eq.17 to obtain our estimates of V for these conditions. Note that while g_{ee} and g_{ei} are in excellent agreement, g_{ii} is considerably more structured in our MD. At first blush, one might expect such a result, since the bare ion-ion plasma coupling is quite large here (~ 12.5), while T_e is possibly high enough to render the electrons nearly equivalent to a negative uniform compensating background which is incapable of affecting much in the way of screening. This last statement is borne out in Fig.7, showing the evolution of the 1st peak in $g_{ii}(r)$ from MD; as T_e is raised from 10^7 K to 10^8 K, the behavior seems to asymptote to

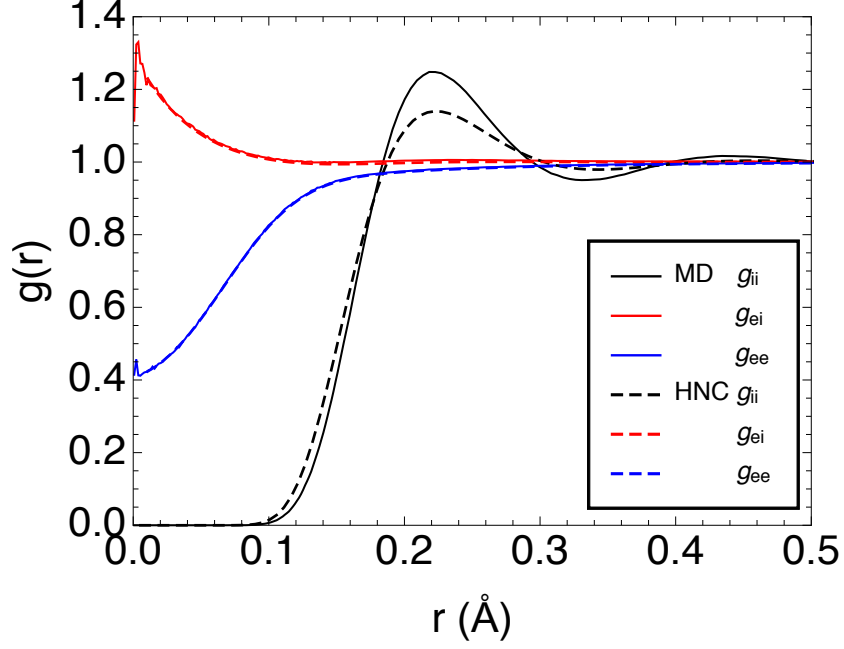


FIG. 6: Plots of $g(r)$ from MD and HNC for the $T_e = 10^7$ K case.

what we must assume is the $T_e \rightarrow \infty$ ion one-component plasma (OCP) result. However, since the $T_e = 10^7$ K g_{ii} [MD] is already much closer to the $T_e = 5 \times 10^8$ K g_{ii} [MD] than it is to the g_{ii} [HNC] shown in Fig.6, it is possible that our HNC predictions of $g_{ii}(r)$ might be rather inaccurate. Two points are worth noting here: 1. The near-independence of g_{ii} (from the MD) on T_e in this regime suggests that the ion-OCP model is in fact a good one for g_{ii} ; this in turn means that the consequences of the detailed assumptions inherent in the *non-equilibrium 2-T* variant of HNC we employ (see the Abstract and the work of Ref.[46]) are minimized. 2. HNC of the 1-T (here equal to T_{ion}) variety is known to work exceedingly well for the OCP when the plasma coupling is ~ 12.5 and far greater. Thus, we can trust that the discrepancy between g_{ii} [MD] and g_{ii} [HNC] is likely not due to a failure of HNC for the *chosen* values of n and T_i .

We therefore explore the possibility that the actual ion-ion correlations in the MD for this case are not those of an ion-OCP with $T_i = 10^5$ K. Fig.8 shows g_{ii} [MD] once again (solid blue curve), together with an ion-OCP $g_{ii}(r)$ as computed with HNC, but at a lower $T_i = 6.777 \times 10^4$ K (solid green curve). They are nearly identical, suggesting that the ions in the non-equilibrium MD calculation are spatially correlated in a manner notably different from that expected from a system in thermal equilibrium at $T_i = 10^5$ K. Indeed,

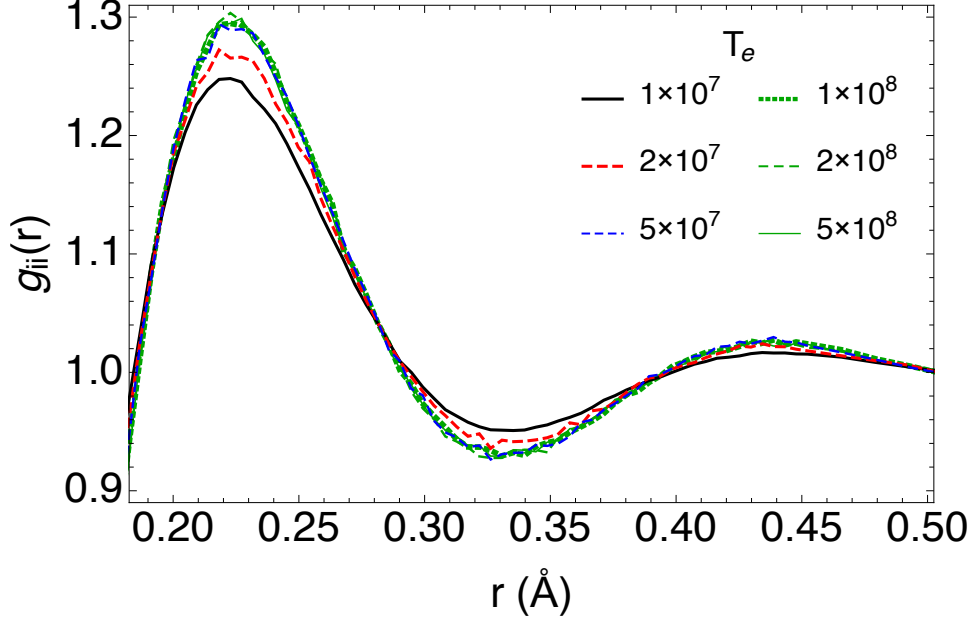


FIG. 7: Close-up of the ion-ion pair distribution functions, for $T_i = 10^5$ K and a series of electron temperatures.

our nominal value of $T_i = 10^5$ K is computed from the ion kinetic energies. We thus find the *kinetic temperature* to be different from the *configurational temperature* here. While we have seen that this has only a modest affect on the analysis of time-varying potential energy in this regime (vis a vis the various T -derivatives of V) [50], we are open to the possibility that such a difference could alter an estimation of the energy exchange rate. As of this writing, we do not have a theoretical tool to assess this, nor do we have a detailed understanding of how, in practice, a sizable difference between kinetic and configurational temperatures could come about. To at least establish that this difference arises from non-adiabatic effects, we have conducted MD studies for this same plasma, but with the proton mass increased by a factor of 8 relative to its physical value. The results for $g_{ii}(r)$ are shown as the black dashed curve in Fig.8. Note that this $m_i = 8m_p$ plasma is less structured than the $m_i = m_p$ plasma; HNC calculations for the ion OCP (not shown) at this density match the black dashed curve well when $T_{\text{config}} \sim 7.5 \times 10^4$ K, intermediate between T_{kinetic} and T_{config} for the associated plasma with the physical ion mass. It is therefore clear that the fundamental source of $T_{\text{config}} \neq T_{\text{kinetic}}$ is the dynamics of the energy exchange process itself. This interesting subject awaits further study.

We have demonstrated that the asymmetric initial relaxations indicated in Figs. 2 and 5

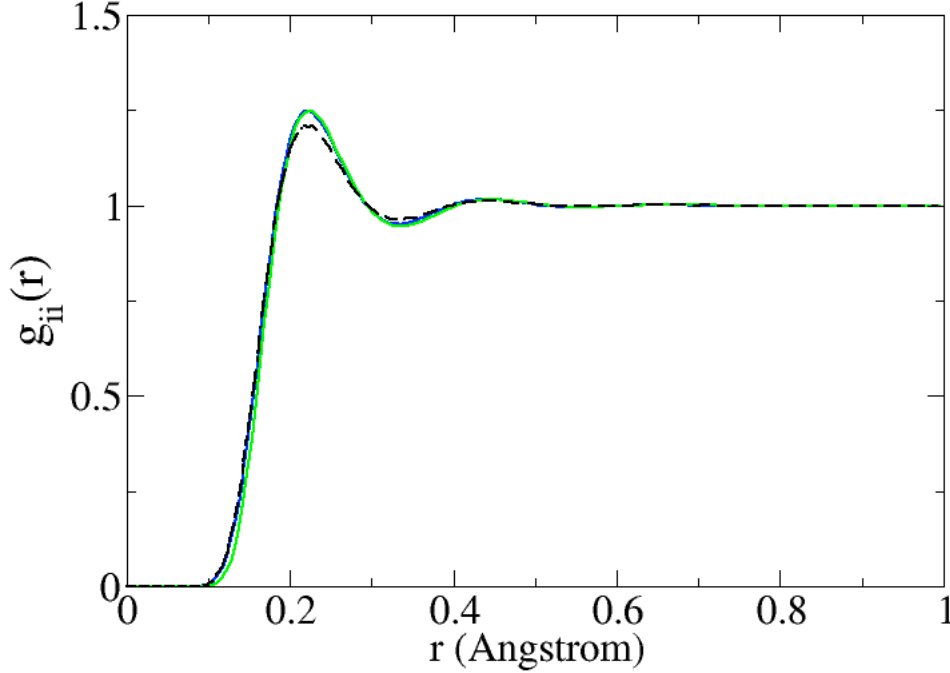


FIG. 8: The instantaneous MD ion-ion pair distribution function, $g_{ii}(r)$, for the hydrogen plasma with $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, and $T_e = 10^7 \text{ K}$ (solid blue curve; essentially obscured by the green curve), compared with the ion OCP $g_{ii}(r)$ for the identical n , but with $T_i = 6.777 \times 10^4 \text{ K}$ (solid green curve). Also shown is the MD result for the same plasma, but with the mass of the protons artificially increased by a factor of 8 (dashed black curve).

result from the T_i -dependence of the potential energy in our classical QSP-MD simulations for hydrogen. This is not surprising, since T_i (10^5 K) is quite low for all of these cases, particularly for such a high density (10^{26} cm^{-3}). However, since these conclusions are based on a classical-QSP model of hydrogen, it is natural to ask: Would such an asymmetry be present in a real hydrogen plasma in these conditions? We address this by adapting the above theoretical estimate, pertaining to Eq.17, to the quantum-electron case by mapping onto an effective one-component (ion-only) Yukawa system as in the work of Ref.[45]. HNC is used once again to compute $g_{ii}(r)$, this time interacting via screened interactions determined from linear response. The electronic part of the free energy is taken from the fit of Tanaka and Ichimaru [51]. Details of our procedure are left to the Appendix. This produces potential

energy derivatives which are broadly similar to those quoted above, $\left(\frac{\partial V}{\partial T_i}\right)_{T_e} = 0.4351 k_B/e^-$, $\left(\frac{\partial V}{\partial T_e}\right)_{T_i} = 0.02868 k_B/e^-$, and derivatives of the total energy (including the quantum electron kinetic energy terms) of $\left(\frac{\partial E}{\partial T_i}\right)_{T_e} = 1.7176 k_B/e^-$, $\left(\frac{\partial E}{\partial T_e}\right)_{T_i} = 1.4347 k_B/e^-$, indicating a $\sim 20\%$ asymmetry between $-dT_e/dt$ and dT_i/dt [52].

Orbital-Free Density Functional Theory (OF-DFT) [53–55] provides another way for us estimate the energetics of 2- T quantum hydrogen plasmas. We employ a treatment that invokes the Born-Oppenheimer approximation, which decouples the electronic and nuclear motions. The nuclei evolve classically, and the electronic contribution derives from a minimization of a quantum free-energy functional in terms of the full electron probability density. We employ a modified Thomas-Fermi-Dirac form that includes the Perrot finite-temperature kinetic-entropic contribution and a local-density exchange-correlation component. The Born-Oppenheimer dichotomy does not permit the direct examination of nonadiabatic processes such as electron-ion equilibration. However, since the OF-DFT MD allows setting T_e and T_i independently for a given simulation, we can elicit the temperature dependence of the total internal energy within this adiabatic scheme. For these comparisons, we consider 432 H atoms (N_i) in a periodic box for trajectories of 10^4 time steps of length 0.012 fs. We have also tested convergence in atom number (128 to 1024), time step, and trajectory length and found variations in the total internal energy E and pressure of less than 1%. By using a range of closely-spaced T_i for a fixed T_e , we find $\left(\frac{\partial E}{\partial T_i}\right)_{T_e} = 2.07 \pm 0.2 k_B/e^-$. As in the above HNC estimates, the variation of E with T_i at fixed T_e is sizable. We therefore expect that a real hydrogen plasma under these conditions will also exhibit an initial asymmetric T -relaxation.

As we mentioned in the Introduction, our main reason for studying out-of-equilibrium plasmas in this manner is to assess and improve upon models currently used in continuum simulations of, e.g., ICF[1, 2]. In such simulations, equation of state (EOS) models are invoked which typically arise from single-temperature constructs which assume that all thermodynamic functions are derived from Helmholtz free energies of the form [57]:

$$F(\rho, T) = E_0(\rho) + F_i(\rho, T) + F_e(\rho, T). \quad (18)$$

Here, E_0 is the "cold" piece representing the density-dependence of the energy of ions fixed in position ($= 0$ for a low-density gas). F_i represents the free energy due to ionic motion (phonons in a solid; more general ionic excitations in a liquid). F_e represents the free energy

of electronic excitations. Implicit is the assumption that ionic and electronic excitations are decoupled; this neglects, for instance, any change in inter-ionic forces that may occur when electrons are strongly excited. When electron-ion T -splits are invoked in most ICF simulations [1], the following approximation is often made to construct a "2- T free energy" from the 1- T free energy above [57]:

$$F_{2T}(\rho, T_i, T_e) \equiv E_0(\rho) + F_i(\rho, T_i) + F_e(\rho, T_e), \quad (19)$$

leading immediately to

$$E_{2T}(\rho, T_i, T_e) = E_0(\rho) + E_i(\rho, T_i) + E_e(\rho, T_e). \quad (20)$$

This assumption of Eq.19 is only reasonable when the EOS itself is practically ideal gas-like. When T_e and T_i are very different *and* the material is somewhat strongly-coupled, it is highly suspect.

Applying this to T -equilibration, the analysis leading to Eq.14, together with the assumption of Eq.20, gives

$$\frac{\left| \frac{dT_e}{dt} \right|}{\left| \frac{dT_i}{dt} \right|} = \frac{\left(\frac{\partial E_i}{\partial T_i} \right)_\rho}{\left(\frac{\partial E_e}{\partial T_e} \right)_\rho} \equiv \frac{C_\rho^i}{C_\rho^e}, \quad (21)$$

the ratio of ionic to electronic specific heats, each evaluated at their respective (e, i) temperatures. For the hydrogen case studied in detail above, we have verified that this prescription produces $C_\rho^i(T_i = 10^5 \text{K}) \sim C_\rho^e(T_e = 10^7 \text{K}) \sim \frac{3}{2} k_B / e^-$, and therefore $\frac{C_\rho^i}{C_\rho^e} \approx 1$, when standard EOS models for hydrogen are used [58]. Thus, such an implementation would fail to predict an initial asymmetric T -equilibration in this case. The essential missing element is the reduced electron screening (and hence increased effective ion-ion interaction) resulting from $T_e \gg T_i$ which is not captured in a treatment assuming $F_i(\rho, T_i, T_e) = F_i(\rho, T_i)$, for which the screened ion-ion interaction is assumed to be derived from a $T_e = T_i$ or $T_e = 0$ theory. This highlights the importance of improving both the in-line models of the equilibration rate, and the EOS model to include more sensible 2- T effects at least at the adiabatic level treated, e.g., in our HNC and OF-DFT calculations.

C. Energy relaxation versus temperature relaxation

As we mentioned when discussing Fig.5, the energy relaxation rate (here multiplied by 2/3 to compare it to temperature relaxation) is quite a bit higher than that as predicted by

Lenard-Balescu using the full coupled-mode treatment. And in fact, it is rather closer to the predictions of FGR, in which coupled-modes are neglected, in contrast to the conclusions of Ref.[20] for these same plasmas. We now discuss this further.

The definitions we use for the internal energies of electrons and ions, respectively, are those of Eq.16. Since $E_e + E_i = E_{\text{total}}$, it follows from energy conservation that $dE_i/dt = -dE_e/dt$. It is worth noting that this is true for *any* definition in which V_{ei} is split between electrons and ions, not just the 50%-50% split. As we have seen from the previous discussion, this perfect asymmetry is not enjoyed by dT_i/dt and dT_e/dt , due to the time-dependence of the total V . We have picked this particular split of V_{ei} in the internal energies because the authors of Ref.[21] argue that this choice alone makes their general expression for the energy equilibration rate (Eq.27, and the more approximate Eq.37 in Ref.[21]) look most like the one derived for weakly-coupled plasmas [59] (Eq.3 in this work, or Eq.7 in Ref.[20]); if the response functions of the more general expressions are replaced by the RPA response functions, the weakly-coupled result is obtained.

The lack of agreement between this expression and the $dE_{e,i}/dt$ from MD for these cases indicates that the response functions at the RPA level are not sufficient. This too was anticipated in the work of Ref.[21], wherein (for hydrogen plasmas with equal densities, but 10 times lower T_i) the inclusion of static ion-ion local field corrections (LFCs) derived from HNC was shown to increase the predicted equilibration rate above that of the RPA CM prediction (see black and red curves in Fig.1 of Ref.[21]). Our analogous predictions appear as the isolated black diamond in Fig.5; only a very slight increase above the RPA CM-LB result (solid black curve and circles) is seen, and the agreement with the energy equilibration rate from MD is therefore still quite poor. Inclusion of all the pairs of static LFCs, G_{ee} , G_{ei} , and G_{ii} , within the dielectric function of Eq.4 produces almost identical results. We are left to conclude that the standard theories for energy equilibration in 2- T plasmas do not apply to at least the semiclassical model of the hydrogen plasmas considered in Ref.[20]. This is troubling for researchers bent on constructing and validating models for energy and temperature equilibration in plasmas with strongly-coupled ions [23, 24]. In the next subsection, we suggest a possible reason for the inability of the CM Lenard-Balescu theory (with RPA screening and/or with static LFCs) to correctly describe the cases of our study; we propose that a variant of *dynamic* local field corrections may be needed here instead. It is worth noting that even in more weakly-coupled regimes, our earlier T -equilibration MD

vs. LB comparisons also showed the inclusion of *static* LFCs to be inadequate (in that case, worse than the RPA result) for oppositely-charged 2-component plasmas [11].

As seen in Fig.5, there are two conditions which are *approximately* satisfied for all the plasmas studied here (if we use the CM-LB results, specifically): 1. $dT_i/dt[\text{MD}] \approx dT_i/dt[\text{LB}]$ ($= 2/3 \times dE_{e,i}/dt[\text{LB}]$), and 2. $-dT_e/dt[\text{MD}] \approx 2/3 \times dE_{e,i}/dt[\text{MD}]$. We currently have no understanding as to why these relations are roughly satisfied, however it is interesting to note that the simultaneous satisfaction of these conditions puts constraints on the 2- T EOS, as expressed in the internal energies of Eq.16. If we assume that

$$E_e = E_e(T_e, T_i),$$

and

$$E_i = E_i(T_e, T_i)$$

[as in our above assumption, $V = V(T_e, T_i)$], application of the chain rule, together with total energy conservation, produces the equations [21]:

$$\begin{aligned} \frac{dE_e}{dt} &\equiv -Z_{\text{MD}} = \left(\frac{\partial E_e}{\partial T_e} \right)_{T_i} \frac{dT_e}{dt} + \left(\frac{\partial E_e}{\partial T_i} \right)_{T_e} \frac{dT_i}{dt}, \\ \frac{dE_i}{dt} &\equiv Z_{\text{MD}} = \left(\frac{\partial E_i}{\partial T_e} \right)_{T_i} \frac{dT_e}{dt} + \left(\frac{\partial E_i}{\partial T_i} \right)_{T_e} \frac{dT_i}{dt}. \end{aligned} \quad (22)$$

Conditions 1 and 2 above can be expressed as: 1. $dT_i/dt[\text{MD}] \approx dT_i/dt[\text{LB}] = 2/3 \times Z_{\text{LB}}$, and 2. $dT_e/dt[\text{MD}] \approx -2/3 \times dE_i/dt[\text{MD}] = -2/3 \times Z_{\text{MD}}$. Plugging these conditions into Eqs.22 and eliminating the ratio $Z_{\text{MD}}/Z_{\text{LB}}$ yields:

$$\left(\frac{\partial E_e}{\partial T_i} \right)_{T_e} \left[\frac{3}{2} + \left(\frac{\partial E_i}{\partial T_e} \right)_{T_i} \right] \approx - \left(\frac{\partial E_i}{\partial T_i} \right)_{T_e} \left[\left(\frac{\partial E_e}{\partial T_e} \right)_{T_i} - \frac{3}{2} \right]. \quad (23)$$

As a concrete example, our HNC estimations, a la Eq.17, for the $T_i = 10^5$ K, $T_e = 10^7$ K case give for this relation (all numbers expressed in units of k_B),

$$-0.043 \times \left[\frac{3}{2} + 0.0011 \right] \approx -2.08 \times \left[1.53 - \frac{3}{2} \right],$$

or

$$-0.0645 \approx -0.0624,$$

demonstrating once again that our HNC-based theoretical modeling of the non-ideal 2- T EOS is on solid ground [60]. More fundamentally, the relative magnitudes of the various

derivatives in Eqs.22 and 23 suggest that the much simpler diagonal form of Eq.22 is quite accurate for these plasmas,

$$\begin{aligned}\frac{dE_e}{dt} &\equiv -Z_{\text{MD}} \approx \left(\frac{\partial E_e}{\partial T_e} \right)_{T_i} \frac{dT_e}{dt} \approx \frac{3}{2} \frac{dT_e}{dt}, \\ \frac{dE_i}{dt} &\equiv Z_{\text{MD}} \approx \left(\frac{\partial E_i}{\partial T_i} \right)_{T_e} \frac{dT_i}{dt} \approx 2 \frac{dT_i}{dt}.\end{aligned}\tag{24}$$

While these observations are clearly borne out in our MD results, we do not claim to understand their origin in a deep sense, beyond the trivial fact that derivatives with respect to T_e are much smaller than derivatives with respect to T_i , due to the small ratio of electronic to ionic coupling. Furthermore, their satisfaction does not *explain* the surprisingly large $Z_{\text{MD}}/Z_{\text{LB}}$ that we have found.

The bare ion-ion plasma coupling for the cases studied heretofore is $\Gamma_{ii} \sim 12.5$. While we have argued that this is low enough for an accurate treatment of static correlations within the rubric of HNC, it is possibly too high for a treatment of the equilibration rates using Eq.3 to be accurate. With this in mind, we also consider the semiclassical hydrogen plasma with $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^6 \text{ K}$, and $T_e = 10^8 \text{ K}$. This possesses a bare $\Gamma_{ii} \sim 1.25$, but with a ratio, T_e/T_i , equal to that of the $T_i = 10^5 \text{ K}$, $T_e = 10^7 \text{ K}$ case. Table III shows our results. Once again, the energy equilibration rate from MD (expressed as an equivalent temperature derivative: 9.97 eV/fs per k_B) is closer to the FGR rate (10.36 eV/fs) than to the CM-LB rate (8.81 eV/fs). And once again, the two approximate conditions discussed directly above are roughly satisfied. Though the difference between dT_i/dt and $-dT_e/dt$ is far smaller in absolute magnitude due to the smaller ion coupling here, the treatment of Ref.[20] and our Eq.3 is still sorely incomplete. It remains to be seen if any plasma studied with non-equilibrium classical MD exhibits the coupled-mode effect in the manner predicted by classical Lenard-Balescu theory with RPA screening.

D. Effects of collisional broadening

As discussed in Ref.[20] and in Section III of this work, the low-energy, long-wave length ion acoustic wave feature in the CM-LB treatment is responsible for the marked reduction in equilibration rates relative to the FGR prediction. Furthermore, this excitation is predicted to be exceedingly sharp in ω (i.e., long-lived in time), within the standard RPA treatment

LB dT_i/dt (no LFCs)	FGR dT_i/dt (no LFCs)	dT_i/dt from MD	$-dT_e/dt$ from MD	$(2/3) \times dE_i/dt$ from MD
8.81	10.36	8.73	9.81	9.97

TABLE III: Instantaneous dT/dt (in eV/fs) calculated from classical-QSP Lenard-Balescu, and extracted from MD using the identical QSPs for a hydrogen plasma with $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^6 \text{ K}$, and $T_e = 10^8$. Also shown in the rightmost column is the instantaneous rate of change of the internal energy of the ions, expressed as $\frac{2}{3}dE_i/dt$, to compare it to the temperature derivatives (Note that the relation $dE_e/dt = -dE_i/dt$ holds exactly, so we only display dE_i/dt).

for the 2-component dielectric function which forms the denominator of Eq.3. It is important to note, however, that MD studies of one- and two-component plasmas in equilibrium have shown that lifetimes of collective modes are greatly reduced relative to those of RPA when the plasma is strongly-coupled. For example, the works of Refs.[61–64] demonstrate large deviations of dynamic structure factors, $S(k, \omega)$, from their RPA predictions in the neighborhood of plasmon peaks. Specifically, these peaks are substantially broadened and reduced in height when the plasma coupling, Γ , is of order a few (see, e.g., Fig.2c and Fig.2d in Ref[62]). Though the static LFCs we have employed in our LB calculations do modify the RPA peak structure in $S(k, \omega)$, they do so primarily by moving the pole positions, rather than by broadening these peaks in frequency. It is therefore of interest to know what effect such a broadening might have on our predictions of T -equilibration.

To wit, we employ a simple scheme for affecting such a broadening, that of replacing $\omega \rightarrow \omega + i\gamma_i$ in χ_i^0 , the density response function of the ions. Here, γ_i represents a phenomenological inverse collisional lifetime. Since this replacement in the Lindhard response of Eq.6 is known to violate local charge conservation, we use instead the modified expression for this function [65] which appears in Refs.[66] and [67]. This new function, $\tilde{\chi}_i^0(k, \omega + i\gamma_i)$, is then input into both the numerator $[\text{Im}\chi_i^0(k, \omega)]$ and the denominator $[D(k, \omega)]$ of Eq.3, the CM-LB expression for dT_i/dt . The electron response function, $\chi_e^0(k, \omega)$, is left unchanged in this expression, because the electrons are far more weakly-coupled in the cases we consider here ($T_e \gg T_i$; $\Gamma_{ee} \sim 0.125$ or less).

Figure 9 shows a series of $F(k)$ k -integrands for the $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, $T_e = 10^7 \text{ K}$ quantum-Coulomb hydrogen case, to be compared to those of Fig.3 above. All have been generated with the aforementioned prescription in which $\tilde{\chi}_i^0(k, \omega + i\gamma_i)$ is used in the context

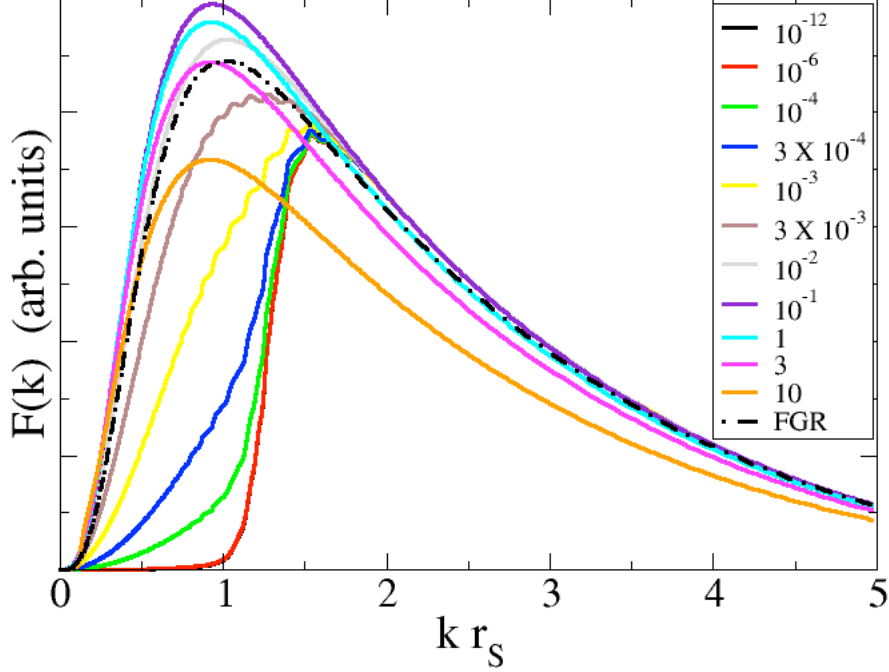


FIG. 9: The k -integrands, $F(k)$, of Eq.3 for the quantum-Coulomb hydrogen plasma with $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, and $T_e = 10^7 \text{ K}$. Here, the Lindhard $\chi_i^0(k, \omega)$ is replaced by the corresponding function, $\tilde{\chi}_i^0(k, \omega + i\gamma_i)$, of Ref.[66, 67]. Each curve represents a different choice of γ_i , expressed in atomic units (Hartrees; see figure key). Note that the $F(k)$ for $\gamma_i = 10^{-12} \text{ Ha}$ (solid black curve; not seen) and the $F(k)$ for $\gamma_i = 10^{-6} \text{ Ha}$ (red curve) are essentially identical.

of Eq.3. Thirteen orders of magnitude in γ_i (in atomic units, Hartrees) are represented. There is a clutch of curves with $10^{-3} < \gamma_i < 10$ located close to the FGR $F(k)$ (black dot-dashed curve), and a collection of curves with $\gamma_i \ll 10^{-3}$ which are essentially indistinguishable from each other and from the RPA CM-LB prediction. Figure 10 shows the integrals under these curves, $\int_0^\infty dk F(k)$ (which are equal to the effective Coulomb-logarithm for this case) as a function of γ_i/ω_p , where ω_p is the ion plasma frequency. The y-axis value of ~ 0.65 denotes the effective $\ln \lambda_{ei}$ corresponding to the CM-LB prediction, while the FGR value is in the neighborhood of $\ln \lambda_{ei} = 0.95$. Thus, it appears that the phenomenological collisional broadening applied solely to the strongly-coupled ions can indeed extinguish the CM effect, *if the collision frequency is in the appropriate range*.

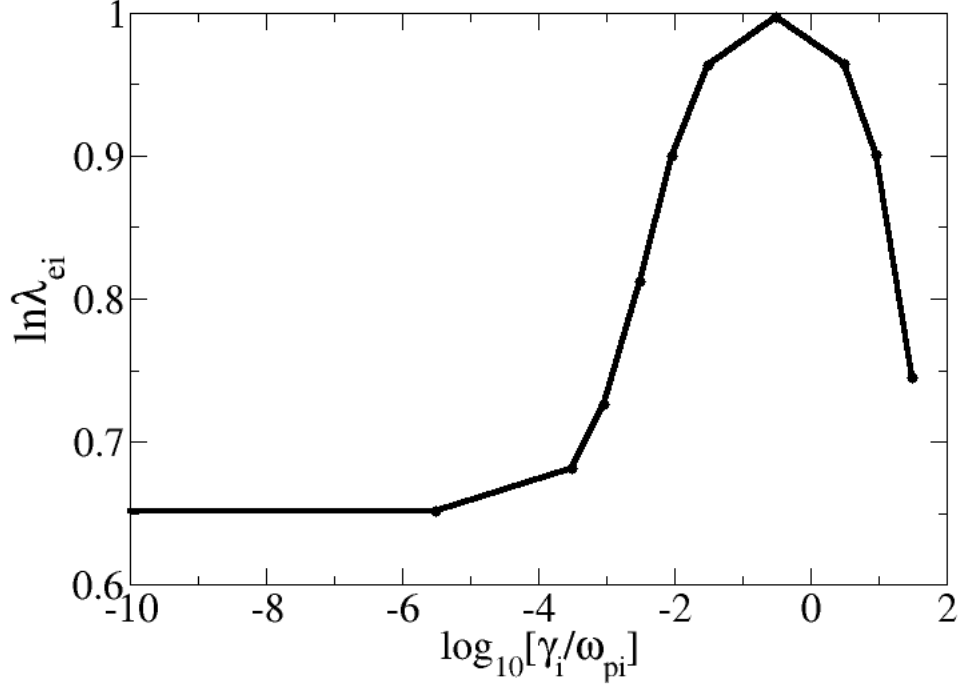


FIG. 10: The effective Coulomb-logarithms, $\ln\lambda_{ei}$, for the quantum-Coulomb hydrogen plasma with $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, and $T_e = 10^7 \text{ K}$. Here, the Lindhard $\chi_i^0(k, \omega)$ in Eq.3, is replaced by the corresponding function, $\tilde{\chi}_i^0(k, \omega + i\gamma_i)$, of Ref.[66, 67]. Each point on the curve represents a different choice of γ_i , expressed in atomic units (Hartrees). The CM-LB RPA result is represented by the nearly flat line at the lower-left of the plot; the FGR result is at $\ln\lambda_{ei} \sim 0.95$.

As an independent estimate of this ion collision frequency, we turn to MD simulations of ion OCPs with plasma conditions similar to ours in this case ($\Gamma \sim 12.5$). The work of Ref.[61] displays MD data for $S(k, \omega)$ for a plasma with $\Gamma = 9.7$ (see their Fig.5). We are concerned primarily with k values of the order of $\sim 1/r_S$, given our discussion surrounding our Fig.3. Both $k = 0.6187/r_S$ and $k = 1.3837/r_S$ were considered in Ref.[61]. To affect a comparison, we evaluate $\tilde{\chi}_i^0(k, \omega + i\gamma_i)$ for the $\Gamma = 9.7$ OCP at these specific k , and for a range of γ_i . From these, we compute the interacting $\tilde{\chi}_i$ functions (which would correspond to the RPA χ in the limit of $\gamma_i \rightarrow 0$), using the usual relation,

$$\tilde{\chi}_i(k, \omega) = \frac{\tilde{\chi}_i^0(k, \omega)}{1 - v(k)\tilde{\chi}_i^0(k, \omega)}. \quad (25)$$

The dynamic structure factors are then computed from these polarizabilities using the

fluctuation-dissipation theorem [39]. Good agreement with the MD results of Ref.[61] are found when γ_i is chosen to be roughly $(0.1 - 0.2) \times \omega_p$. The plot of Fig.10 shows this to be in the range where the FGR result is expected. Yet another estimate for γ_i can be obtained by appealing to the relationship between this quantity and the self-diffusion constant, D , outlined in chapter 9 of Ref.[68] and alluded to in Ref.[69]:

$$\gamma = \frac{k_B T}{mD}, \quad (26)$$

where m is the ion mass. Using the diffusion constant for the $\Gamma = 9.7$ plasma quoted in Table II of Ref.[61], we obtain $\gamma_i = 0.26 \times \omega_p$, once again in the range clearly favoring the FGR result. Finally, our own MD computation of $S(k = 0.3/r_S, \omega)$ for the $\Gamma = 12.5$ OCP shows a plasmon peak with a full-width at half maximum of $\sim 0.1 \times \omega_p$, again indicating a γ_i in this same range.

We therefore deem it highly likely that collisional broadening among the strongly-coupled ions is responsible for extinguishing the coupled-mode effect. It may prove fruitful in future studies to explore more complex k -dependent models for γ_i [63, 64] as well as more sophisticated (beyond-RPA) treatments of the plasma dispersion relation to refine these predictions of $S_{ii}(k, \omega)$ further, as they apply to the equilibration problem. More fundamentally however, it is unknown at present if the GLB prescription, as exhibited in Eq.3, is truly up to the task of handling such problems; our lifetime-broadened $S_{ii}(k, \omega)$ is essentially equivalent to some specific dynamical local-field correction $[G_{ii}(k, \omega)]$ as appearing in Eqs.3 and 4, but for sufficiently strong plasma couplings, even this picture is likely to be inadequate for readily accessible $G(k, \omega)$.

E. Effect of non-Maxwellian velocity distributions

Because the plasmas we consider here have such large initial T -splits, and the cases we have studied exhibit distinct kinetic and configurational ion temperatures, it is of interest to know if the assumption of Maxwellian velocity distributions (which we have made thus far in the theoretical estimates) is truly well-founded for the time intervals over which MD data has been taken. It is conceivable that the disagreements we have seen, LB vs. MD, result in part from the inadequacy of the Maxwellian assumption. To this end, we examine the velocity distributions from MD, $f_e(v)$ and $f_i(v)$, at representative times for the trajectory

corresponding to the $n = 10^{26} \text{ cm}^{-3}$, $T_i = 10^5 \text{ K}$, $T_e = 10^7 \text{ K}$ case. Figure 11 shows the ion distribution (purple) from our MD simulation, together with the Maxwellian (green) at the T_i corresponding to the total ion kinetic energy at this t . A small but systematic deviation between the two is noticeable. Figure 12 shows the residual (purple curve minus green curve in Fig.11) over this same energy range (purple).

The green curve in Fig.12 is our smooth fit to this residual, constructed by expanding the multiplicative deviation of $f_i(v)$ from the Maxwellian, $f_i^{(0)}(v)$, in a series of Laguerre polynomials,

$$f_i(v) = f_i^{(0)}(v) \sum_{n=0}^{\infty} A_n^i L_n^{(1/2)} \left(\frac{m_i \beta_i v^2}{2} \right). \quad (27)$$

We find that the residual is best fit with *just the first non-unity term* in this expansion. The cyan curve in Fig.11 shows $f_i^{(0)}(v)$ plus our fit to the residual. The electron distribution at this same time (and for all times) has practically no residual at all; this is to be expected from the fact that the ions move much more slowly than the electrons and are therefore slower to come into intra-species equilibrium [71].

Using the mathematical techniques employed in Ref.[72], we predict that the ion residual shown in Fig.12 increases the LB prediction of dT_i/dt by only $\sim 3 \%$, relative to that of the perfect Maxwellian with $T = T_i$. Details of this calculation are given in the Appendix. This then demonstrates that *intraspecies* equilibration is indeed much faster than the timescale for interspecies heat exchange of primary interest in this work, as expected. Thus, despite the large initial T -splits which could in principle have skewed the MD velocity distributions appreciably at later times, we conclude that the plasmas we study here are indeed amenable to theoretical T -equilibration treatments which assume Maxwellian velocity distributions. However, the failure of the ion static structure [represented by $g_{ii}(r)$] to reflect the correlations of a system of ions in thermal equilibrium at the kinetic temperature, T_i , is another matter entirely, and is undoubtedly a manifestation of non-adiabatic effects which are beyond the scope of our current theoretical tools (a la Eq.3).

V. CONCLUSIONS

We have presented non-equilibrium classical molecular dynamics results for temperature equilibration of hydrogen plasmas at a density of $n = 10^{26} \text{ cm}^{-3}$. Inter-particle interactions were taken to be quantum statistical potentials (QSP) of the Dunn-Broyles [27] and Minoo

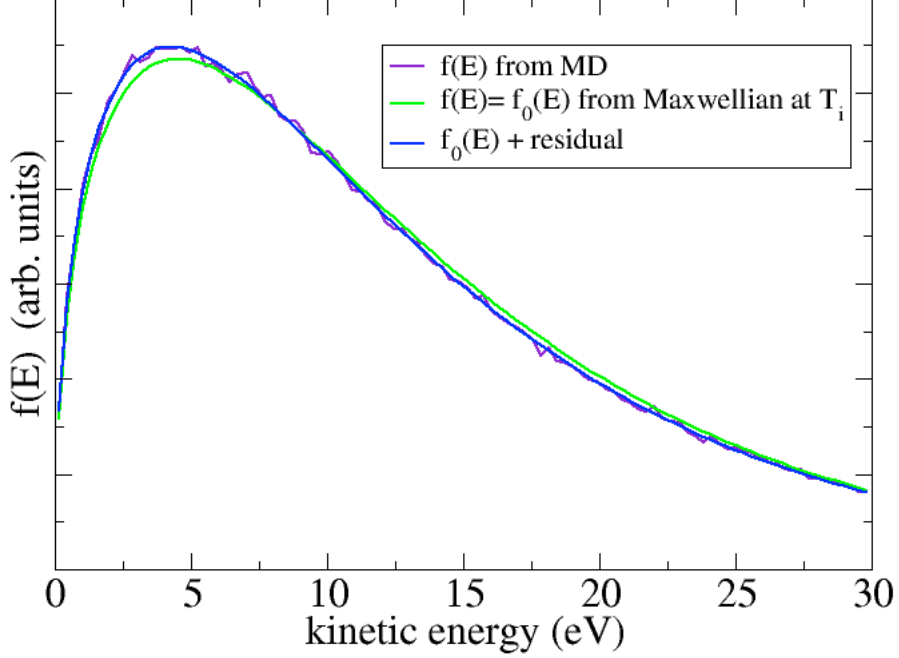


FIG. 11: ion kinetic energy distributions from MD (purple), together with the closest-fit Maxwellian.

et al. [28] forms [35]. The initial temperatures considered, $T_i = 10^5$ K and T_e between 10^7 K and 5×10^8 K, were chosen to coincide with those studied in a paper of Vorberger and Gericke [20] in which they used the Lenard-Balescu equation to predict that the energy equilibration rate for such systems is substantially lowered due to the presence of ion acoustic waves. Our Lenard-Balescu calculations for the associated classical-QSP plasmas also predict a corresponding reduction, but fail to agree with the energy equilibration rates from our MD. Inclusion of static local-field corrections in the (generalized) Lenard-Balescu calculations does little to improve this. However, replacing the Lindhard ion density response function, χ_i^0 , by one in which collisional broadening is assumed largely extinguishes the coupled-mode effect.

The large differences between T_e and T_i complicate the preparation of non-equilibrium systems compared to what has been studied the past [11, 13]. Thus, we discussed the relative merits of three schemes for initializing the MD simulations. The evaluation of the

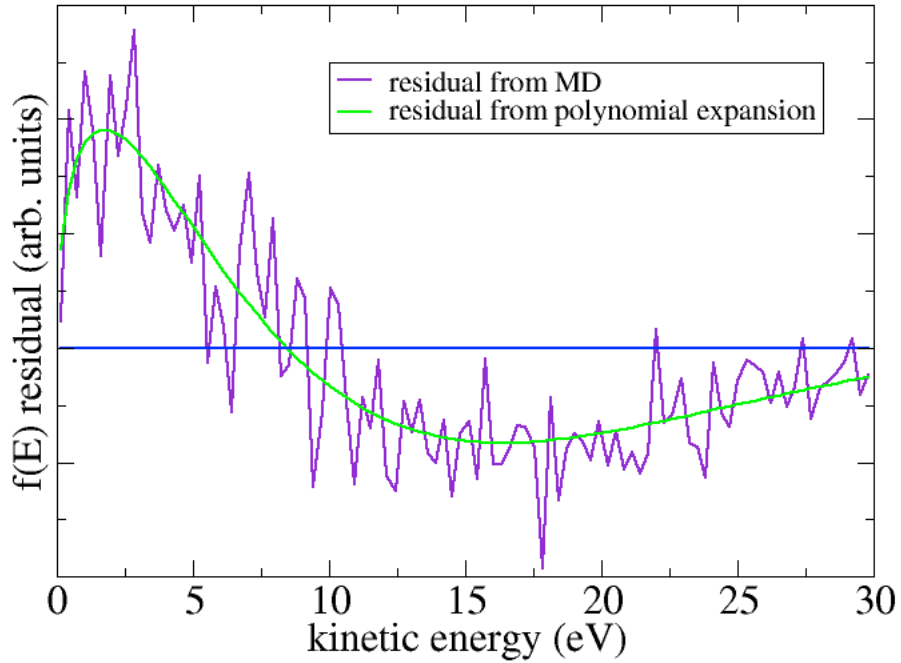


FIG. 12: The KE_{ion} -dependent residual, together with a fit comprised (chiefly) a single Laguerre polynomial.

Lenard-Balescu equilibration rate is also more complex when the plasmas considered are in this regime; our strategy for computing it was described, and comparisons were made to the analogous strategies and results outlined in Ref.[20].

We found that for the cases we studied with MD, there is a sizable effect on dT_e/dt and dT_i/dt from time-varying potential energy, not treated in the usual Lenard-Balescu approach, but discussed in detail in various recent works [7, 14, 21]. We used a variety of approaches to quantify this effect, and we also demonstrated that it should be present for real quantum hydrogen plasmas in these conditions as well (i.e., not just for the classical-QSP model of hydrogen with which we have conducted our MD simulations of T -equilibration). In addition, we observed that the static structure of the ions in our classical MD simulations is notably different (more structured) from that corresponding to ions *in thermal equilibrium* at the instantaneous kinetic- T_i . This may very well result from strong non-adiabatic effects in the systems under consideration, the implications of which must be studied further.

Our MD results and associated comparisons to theory constitute a crucial step in validating and critiquing new models for temperature equilibration [23, 24] aimed at treating plasmas for which the electron-electron and electron-ion couplings may be weak, but the ion-ion coupling is somewhat stronger. In addition to being of potential interest to the field of laser-produced plasmas (where, generally, $T_e \gg T_i$ initially), this sets the stage for the investigation of plasmas containing multiple ion species where some of these ions have a sizable average charge. Such cases have also been predicted to be strongly affected by coupled collective modes (see Eq.1). However, we are now faced with the troubling result that the coupled-mode effect itself seems to be greatly exaggerated in the available theories, at least for the 2-species plasmas we have considered in this work. It is entirely possible that the ion acoustic waves which constitute the coupled modes here are so severely underdamped as to be veritably decoupled from the other modes (e.g., plasmons) in the system; in this case, such modes may not thermalize sufficiently to steal oscillator strength away from the plasmons, as they are assumed to do in the coupled-mode Lenard-Balescu treatment [20]. Still another potential explanation is that stemming from the calculations we have outlined in Section IV D, collisional broadening within the strongly-coupled ion subsystem.

It remains to be seen if a more sophisticated treatment of the coupled electron-ion density response [21] yields more favorable comparisons with MD, or if the linear response assumption itself is questionable in these $T_e \gg T_i$ cases.

VI. ACKNOWLEDGMENTS

We thank D.O. Gericke, J. Vorberger, D.A. Chapman, H.D. Whitley, and A. Pribram-Jones for helpful discussion. 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 by Los Alamos National Laboratory under Contract DE-AC52-06NA25396.

Appendix A: Model for the internal energy of a 2- T hydrogen plasma

To approximate the internal energy of a real Coulomb system, we follow a derivation similar to that given by Ashcroft and Stroud [45]. The general Hamiltonian of a Coulomb

system (classical or quantum) can be written as

$$\mathcal{H} = \sum_i \frac{p_i^2}{2m} + \frac{1}{2} \sum_{i \neq i'} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_{i'}|} - E_0 \quad (\text{A1})$$

$$+ \frac{1}{2} \sum_{j \neq j'} \frac{Z^2 e^2}{|\mathbf{R}_j - \mathbf{R}_{j'}|} + E_1 + E_0 \quad (\text{A2})$$

$$- \sum_{i,j} \frac{Ze^2}{|\mathbf{r}_i - \mathbf{R}_j|} - E_1 \quad (\text{A3})$$

$$+ \sum_j \frac{P_j^2}{2M}, \quad (\text{A4})$$

where n_e is the mean electron number density, E_0 is the self-energy of a uniform background of charge density $n_e e$ given by

$$E_0 = \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \frac{(n_e e)^2}{|\mathbf{r} - \mathbf{r}'|}, \quad (\text{A5})$$

and E_1 is the interaction between the ions and this background

$$E_1 = \frac{1}{2} \sum_j \int d\mathbf{r} \frac{Ze^2 n_e}{|\mathbf{r} - \mathbf{R}_j|}. \quad (\text{A6})$$

Here, $\{\mathbf{r}_i\}$ and $\{\mathbf{R}_j\}$ denote electronic and ionic coordinates respectively with $\{\mathbf{p}_i\}$ and $\{\mathbf{P}_j\}$ being their momenta and m and M being their masses. The lines of (A1-A4) have been arranged to yield components that are individually finite in the thermodynamic limit. For example, line (A1) is simply the Hamiltonian of a free electron gas, \mathcal{H}_{feg} , embedded in a neutralizing uniform background. The next two lines (A2-A3) can be expressed in Fourier space as

$$\mathcal{V}_{ii} = \frac{Z^2}{2} \int \frac{d\mathbf{k}}{(2\pi)^3} v_{ee}(k) [\hat{n}_{\text{ion}}(\mathbf{k}) \hat{n}_{\text{ion}}(-\mathbf{k}) - N_{\text{ion}}], \quad (\text{A7})$$

$$\mathcal{V}_{ei} = -\frac{Z}{2} \int \frac{d\mathbf{k}}{(2\pi)^3} v_{ee}(k) \hat{n}_{\text{ion}}(\mathbf{k}) \hat{n}_e(-\mathbf{k}), \quad (\text{A8})$$

where $\hat{n}_{\text{ion}}(\mathbf{k})$ and $\hat{n}_e(\mathbf{k})$ are the ion and electron density operators respectively, N_{ion} is the total number of ions, and $v_{ee}(k) = 4\pi e^2/k^2$.

Generally speaking, the statistical mechanics of a two-temperature system is poorly defined [44]. However, for the electron-proton system of interest, we can simplify the calculations by approximating the ions as classical point particles and temporarily fixing their coordinates, which is reasonable given their mass discrepancy ($M/m \approx 1836$) and lower

temperature ($T_i/T_e \leq 1/100$). The ensemble average over the electron degrees of freedom in terms of T_e generates the internal energy of the electron gas $\langle \mathcal{H}_{\text{feg}} \rangle = N_{\text{ion}} U_{\text{feg}}(n_e, T_e)$, the electron density $\langle \hat{n}_e(\mathbf{k}) \rangle$ and leaves the remaining variables unchanged. This procedure is equivalent to the Born-Oppenheimer approximation.

We next approximate the electron density in terms of the ion density operator through a response function as

$$\langle \hat{n}_e(\mathbf{k}) \rangle \approx -Z v_{ee}(k) \chi(k; n_e, T_e) n_{\text{ion}}(\mathbf{k}). \quad (\text{A9})$$

In general, this response function would be calculated in terms of cross-correlations between the ions and electrons and thus depend on both temperatures; however, the response will be dominated by the electron temperature for the problem of interest due to the previously mentioned disparate quantities [46, 48]. Next, the ionic coordinates are released, and an ensemble average is taken in terms of the ionic degrees of freedom and T_i . Using the definition of the ion-ion static structure factor

$$S_{ij}(k) \equiv \frac{1}{\sqrt{N_i N_j}} \langle \hat{n}_i(\mathbf{k}) \hat{n}_j(-\mathbf{k}) \rangle, \quad (\text{A10})$$

we obtain the expression for the total internal energy

$$\langle \mathcal{H} \rangle = N_{\text{ion}} U_{\text{int}} \quad (\text{A11})$$

$$= N_{\text{ion}} U_{\text{feg}}(n_e, T_e) \quad (\text{A12})$$

$$+ N_{\text{ion}} \frac{Z^2}{2} \int \frac{d\mathbf{k}}{(2\pi)^3} v_{ee}(k) [S_{ii}(k) - 1] \quad (\text{A13})$$

$$+ N_{\text{ion}} \frac{Z^2}{2} \int \frac{d\mathbf{k}}{(2\pi)^3} v_{ee}^2(k) \chi(k) S_{ii}(k) \quad (\text{A14})$$

$$+ \frac{3}{2} N_{\text{ion}} k_B T_i, \quad (\text{A15})$$

where lines (A12-A15) correspond to lines (A1-A4) respectively. By next introducing the dielectric function $\epsilon^{-1}(k) = 1 + v_{ee}(k) \chi(k)$, we can write the internal energy *per ion* as

$$U_{\text{int}} = U_{\text{feg}} + \frac{3}{2} k_B T_i + \frac{Z^2}{2} \int \frac{d\mathbf{k}}{(2\pi)^3} v_{ee}(k) \left[\frac{S_{ii}(k)}{\epsilon(k)} - 1 \right]. \quad (\text{A16})$$

As a simple model, we take the response function associated with the linearized Thomas-Fermi (TF) functional. Within this approximation, the dielectric function takes the form

$\epsilon(k) \approx 1 + (\lambda_{\text{TF}} k)^{-2}$, where the TF screening length is $\lambda_{\text{TF}}^{-2} = 4\pi e^2 (\partial n_e / \partial \mu)$ with

$$n_e = 2 \int \frac{d\mathbf{p}}{(2\pi)^3} \left(1 + e^{(p^2/2m - \mu)/T_e} \right)^{-1}. \quad (\text{A17})$$

More complicated energy functionals have been explored with gradient corrections to the density and exchange-correlation effects, but these higher-order contributions were found to be negligible for the system of interest [70]. The resulting internal energy then simplifies to

$$U_{\text{int}} = U_{\text{feg}}(n_e, T_e) + \frac{3}{2} k_B T_i - \frac{Z^2}{2\lambda_{\text{TF}}} + 2\pi Z^2 n_{\text{ion}} \int_0^\infty dr e^{-r/\lambda_{\text{TF}}} r [g_{ii}(r) - 1], \quad (\text{A18})$$

where $g_{ii}(r)$ is the ion-ion radial distribution function. Finally, the internal energy of the electron gas can be estimated using fits from Reference [51], while the final potential energy term in (A18) can be calculated using HNC with the appropriate screened Coulomb (Yukawa) interaction.

Appendix B: Equilibration rates for non-Maxwellian distributions

We compute energy-equilibration for a plasma with electrons and one ion species, for the case of general particle distributions which include small non-Maxwellian components, using the quantum Lenard-Balescu equation. Though our primary aim in the bulk of the main text is the analysis of classical plasmas, we use quantum-LB here because the quantum-LB collision operator is mathematically more straightforward to handle in this context. However, we expect the effects of non-Maxwellian distributions on equilibration rates to be largely insensitive to this distinction. Considering two species, electrons and protons, their distributions are written as:

$$f_e(v) = f_e^{(0)}(\mathbf{v}) \sum_{n=0}^{\infty} A_n^e L_n^{(1/2)} \left(\frac{m_e \beta_e v^2}{2} \right), \quad (\text{B1})$$

$$f_i(v) = f_i^{(0)}(\mathbf{v}) \sum_{n=0}^{\infty} A_n^i L_n^{(1/2)} \left(\frac{m_i \beta_i v^2}{2} \right), \quad (\text{B2})$$

where $f_e^{(0)}$ and $f_i^{(0)}$ are Maxwellians, and $L_n^{(1/2)}$ are Laguerre polynomials, which allow for complete flexibility in the shapes of the distributions. Conservation of the number of particles for each species requires [72],

$$A_0^e = A_0^i = 1. \quad (\text{B3})$$

The condition for kinetic energy conservation (this is all we can assume here, given our use of LB [16]) can be derived from

$$KE_e = \frac{3}{2} \frac{n_e}{\beta_e}, KE_i = \frac{3}{2} \frac{n_i}{\beta_i}, \quad (\text{B4})$$

which gives

$$A_1^e = A_1^i = 0. \quad (\text{B5})$$

Since, for hydrogen, we have $n_e = n_i$,

$$\beta_i A_1^e = -\beta_e A_1^i. \quad (\text{B6})$$

The 2-species LB kinetic equation is of the form,

$$\frac{\partial f_e}{\partial t} = C_{ee}(f_e) + C_{ei}(f_e, f_i), \quad (\text{B7})$$

$$\frac{\partial f_i}{\partial t} = C_{ii}(f_i) + C_{ie}(f_e, f_i). \quad (\text{B8})$$

The intra-species collision operators, C_{ee} and C_{ii} , are irrelevant for energy equilibration. We compute the time rate of change of the ion energy,

$$\frac{d(KE)_i}{dt} = \frac{1}{2} m_i \int v^2 C_{ie}(f_e, f_i) d^3 \mathbf{v}. \quad (\text{B9})$$

The collision operator is given by [73]

$$\begin{aligned} C_{ie}(f_e, f_i) = & -\frac{1}{4\pi^2 \hbar^2} \int d^3 \mathbf{v}' \int d^3 \mathbf{k} \frac{|\phi_{ei}(k)|^2}{\left| \epsilon \left(k, \mathbf{k} \cdot \mathbf{v} + \frac{\hbar k^2}{2m_i} \right) \right|^2} \\ & \times \delta[\mathbf{k} \cdot (\mathbf{v} - \mathbf{v}') + \hbar k^2 / 2\mu] \\ & \times [f_i(\mathbf{v}) f_e(\mathbf{v}') - f_i(\mathbf{v} + \hbar \mathbf{k} / m_i) f_e(\mathbf{v}' - \hbar \mathbf{k} / m_e)], \end{aligned} \quad (\text{B10})$$

where

$$\mu \equiv \frac{m_e m_i}{m_e + m_i} \quad (\text{B11})$$

is the reduced mass. Next we plug the distributions of (B1) and (B2) into (B9) and use various identities to perform the integrals. The \mathbf{v}' integrals can be handled immediately,

$$I_1(k, \omega) \equiv \int d^3 \mathbf{v}' \delta(\omega_+ - \mathbf{k} \cdot \mathbf{v}') f_e(\mathbf{v}') =$$

$$n_e \left(\frac{\beta_e m_e}{2\pi} \right)^{1/2} \frac{1}{k} \sum_n A_n^e L_n^{(-\frac{1}{2})}(Y_+^2) e^{-Y_+^2} \quad (\text{B12})$$

$$I_2(k, \omega) \equiv \int d^3 \mathbf{v}' \delta(\omega_+ - \mathbf{k} \cdot \mathbf{v}') f_e(\mathbf{v}' - \hbar \mathbf{k}/m_e) =$$

$$n_e \left(\frac{\beta_e m_e}{2\pi} \right)^{1/2} \frac{1}{k} \sum_n A_n^e L_n^{(-\frac{1}{2})}(Y_-^2) e^{-Y_-^2}, \quad (\text{B13})$$

where

$$\omega \equiv \mathbf{k} \cdot \mathbf{v} + \frac{\hbar k^2}{2m_i}, \quad (\text{B14})$$

$$\omega_{\pm} \equiv \omega \pm \frac{\hbar k^2}{2m_e}, \quad (\text{B15})$$

and

$$Y_{\pm}^2 \equiv \frac{m_e \beta_e \omega_{\pm}^2}{2k^2}. \quad (\text{B16})$$

We then have

$$\begin{aligned} \frac{d(KE)_i}{dt} &= -\frac{1}{4\pi^2 \hbar^2} \frac{1}{2} m_i \int d^3 \mathbf{v} \int d^3 \mathbf{k} \frac{|\phi_{ei}(k)|^2}{|\epsilon(k, \omega)|^2} v^2 \\ &\quad \times [f_i(\mathbf{v}) I_1(k, \omega) - f_i(\mathbf{v} + \hbar \mathbf{k}/m_i) I_2(k, \omega)]. \end{aligned} \quad (\text{B17})$$

Turning to the \mathbf{v} integrals,

$$J_1(k) \equiv \int d^3 \mathbf{v} \frac{|\phi(k)|^2}{|\epsilon(k, \omega)|^2} v^2 f_i(\mathbf{v}) I_1(k, \omega), \quad (\text{B18})$$

$$\begin{aligned} J_2(k) &\equiv \int d^3 \mathbf{v} \frac{|\phi(k)|^2}{|\epsilon(k, \omega)|^2} v^2 f_i(\mathbf{v} + \hbar \mathbf{k}/m_p) \\ &\quad \times I_2(k, \omega). \end{aligned} \quad (\text{B19})$$

For these, we assume the \mathbf{k} vector points in the z -direction so that

$$v_z = \frac{\bar{\omega}_-}{k}, \quad (\text{B20})$$

$$dv_z = \frac{1}{k} d\omega \quad (\text{B21})$$

and

$$v^2 = v_{\perp}^2 + \left(\frac{\bar{\omega}_-}{k} \right)^2, \quad (\text{B22})$$

where

$$\bar{\omega}_{\pm} \equiv \omega \pm \frac{\hbar k^2}{2m_i}. \quad (\text{B23})$$

To evaluate $J_1(k)$ and $J_2(k)$, we need the identities

$$\begin{aligned} \int_0^\infty e^{-\beta_i m_i v_\perp^2/2} L_m^{(1/2)} \left(\frac{\beta_i m_i v_\perp^2}{2} + \frac{\beta_i m_i v_z^2}{2} \right) v_\perp dv_\perp = \\ \frac{1}{\beta_i m_i} L_m^{(-\frac{1}{2})} \left(\frac{\beta_i m_i v_z^2}{2} \right), \end{aligned} \quad (\text{B24})$$

and

$$\begin{aligned} \int_0^\infty e^{-\beta_i m_i v_\perp^2/2} L_m^{(1/2)} \left(\frac{\beta_i m_i v_\perp^2}{2} + \frac{\beta_i m_i v_z^2}{2} \right) v_\perp^3 dv_\perp = \\ \frac{4}{(\beta_i m_i)^2} L_m^{(-\frac{3}{2})} \left(\frac{\beta_i m_i v_z^2}{2} \right). \end{aligned} \quad (\text{B25})$$

Note that

$$\frac{\beta_i m_i v_z^2}{2} = \bar{Y}_-, \quad (\text{B26})$$

where

$$\bar{Y}_- \equiv \frac{\beta_i m_i \bar{\omega}_\pm^2}{2k^2}. \quad (\text{B27})$$

Using these relations, we obtain

$$\begin{aligned} J_1(k) = \sqrt{\frac{2}{\pi}} \frac{n_i}{\sqrt{m_i \beta_i}} \sum_m A_m^i \int_{-\infty}^\infty \frac{|\phi(k)|}{|\epsilon(k, \omega)|^2} e^{-\bar{Y}_-} \\ \times \left[2L_m^{(-\frac{3}{2})} (\bar{Y}_-) + \bar{Y}_- L_m^{(-\frac{1}{2})} (\bar{Y}_-) \right] \frac{I_1(k, \omega)}{k} d\omega. \end{aligned} \quad (\text{B28})$$

To compute $J_2(k)$, we make the substitution $\mathbf{u} = \mathbf{v} + \hbar \mathbf{k}/m_p$. Because we choose \mathbf{k} to point in the z -direction, we have

$$v^2 = u_\perp^2 + \left(\frac{\bar{\omega}_-}{k} \right)^2 \quad (\text{B29})$$

and we can immediately write

$$\begin{aligned} J_2(k) = \sqrt{\frac{2}{\pi}} \frac{n_i}{\sqrt{m_i \beta_i}} \sum_m A_m^i \int_{-\infty}^\infty \frac{|\phi(k)|^2}{|\epsilon(k, \omega)|^2} e^{-\bar{Y}_+} \\ \times \left[2L_m^{(-\frac{3}{2})} (\bar{Y}_+) + \bar{Y}_+ L_m^{(-\frac{1}{2})} (\bar{Y}_+) \right] \frac{I_2(k, \omega)}{k} d\omega. \end{aligned} \quad (\text{B30})$$

The equilibration rate is given by

$$\frac{d(KE)_i}{dt} = -\frac{m_i}{2\pi\hbar^2} \int_0^\infty [J_1(k) - J_2(k)] k^2 dk. \quad (B31)$$

Noting that $I_1(k, \omega) = I_2(k, -\omega)$, we can show by the substitution $\omega \rightarrow -\omega$ that

$$J_2(k) = \sqrt{\frac{2}{\pi}} \frac{n_i}{\sqrt{m_i\beta_i}} \sum_m A_m^i \int_{-\infty}^\infty \frac{|\phi(k)|^2}{|\epsilon(k, \omega)|^2} e^{-\bar{Y}_-^2} \\ \times \left[2L_m^{(-\frac{3}{2})}(\bar{Y}_-^2) + \bar{Y}_+^2 L_m^{(-\frac{1}{2})}(\bar{Y}_-^2) \right] \frac{I_1(k, \omega)}{k} d\omega, \quad (B32)$$

and

$$J_1(k) - J_2(k) = \sqrt{\frac{2}{\pi}} \frac{n_i}{\sqrt{m_i\beta_i}} \sum_m A_m^i \times \int_{-\infty}^\infty \frac{|\phi(k)|^2}{|\epsilon(k, \omega)|^2} e^{-\bar{Y}_-^2} \\ \times \left[(\bar{Y}_-^2 - \bar{Y}_+^2) L_m^{(-\frac{1}{2})}(\bar{Y}_-^2) \right] \frac{I_1(k, \omega)}{k} d\omega. \quad (B33)$$

We now need to plug $I_1(k, \omega)$ into this formula, which we do by making use of the identities

$$\bar{Y}_-^2 - \bar{Y}_+^2 = -\beta_i \hbar \omega, \quad (B34)$$

and

$$\bar{Y}_-^2 + \bar{Y}_+^2 = \frac{m_i\beta_i}{2k^2} (1 + \alpha) \omega^2 + \frac{\hbar\omega}{2} (\beta_e - \beta_i) \\ + \frac{\hbar^2 k^2}{8} \left(\frac{\beta_i}{m_i} + \frac{\beta_e}{m_e} \right), \quad (B35)$$

where

$$\alpha \equiv \frac{m_e\beta_e}{m_i\beta_i}. \quad (B36)$$

Putting these pieces together, we find

$$\frac{d(KE)_i}{dt} = 8n_e n_i \sqrt{\frac{\beta_e}{2m_i}} e^4 \sum_{m=0}^\infty \sum_{n=0}^\infty A_n^e A_m^i \\ \times \int_0^\infty \int_{-\infty}^\infty \frac{e^{-(1+\alpha)x^2}}{|\epsilon(x, y)|^2} \frac{e^{-\sqrt{\frac{m_e}{m_i}} 2xy(\gamma-1)}}{y^2} e^{-(\gamma+m_e/m_i)y^2} \\ \times L_m^{(-\frac{1}{2})}(\bar{Y}_-^2) L_n^{(-\frac{1}{2})}(\bar{Y}_+^2) x dx dy, \quad (B37)$$

where we have used the dimensionless variables

$$x^2 = \frac{m_i \beta_i \omega^2}{2k^2}, \quad y^2 \equiv \frac{\hbar^2 \beta_i k^2}{8m_e}, \quad \gamma \equiv \frac{\beta_e}{\beta_i} = \frac{T_i}{T_e}, \quad (\text{B38})$$

which lead to

$$Y_{\pm} = \sqrt{\alpha}x \pm \sqrt{\gamma}y, \quad (\text{B39})$$

$$\bar{Y}_{\pm} = x \pm \sqrt{\frac{m_e}{m_i}}y. \quad (\text{B40})$$

Now we need the two-component RPA dielectric function for which the distributions of (B1) and (B2) are used. This is derived in Appendix A of Ref.[72, 74]:

$$\epsilon(x, y) = 1 + \frac{\eta^2}{y^2} \left[w^i(x) + \frac{\xi \gamma}{Z^2} w^e(x) \right], \quad (\text{B41})$$

where

$$w_r^i(x) = \sum_k A_k^i M\left(k+1, \frac{1}{2}; -x^2\right), \quad (\text{B42})$$

$$w_i^i(x) = \sqrt{\pi} x e^{-x^2} \sum_k A_k^i L_k^{(-\frac{1}{2})}(x^2), \quad (\text{B43})$$

and

$$w_r^e(x) = \sum_k A_k^e M\left(k+1, \frac{1}{2}; -\alpha x^2\right), \quad (\text{B44})$$

$$w_i^e(x) = \sqrt{\pi} \sqrt{\alpha} x e^{-\alpha x^2} \sum_k A_k^e L_k^{(-\frac{1}{2})}(\alpha x^2). \quad (\text{B45})$$

Here, $M(a, b; z)$ is the confluent hypergeometric function. This result, together with (B37) and the method of Appendix C of Ref.[72], provides us with the means to compute (kinetic) energy equilibration rates for non-Maxwellian particle distributions within LB.

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of Section IV.1 substantively. As explained in Ref.[72], this assumption is made strictly for mathematical convenience.