

Molecular dynamics simulations of temperature equilibration in dense hydrogen

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The temperature equilibration rate between electrons and protons in dense hydrogen has been calculated with molecular dynamics simulations for temperatures between 10 and 600 eV and densities between 10^{20} cm⁻³ to 10^{24} cm⁻³. Careful attention has been devoted to convergence of the simulations, including the role of semiclassical potentials. We find that for Coulomb logarithms $\mathcal{L} \geq 1$, a model by Gericke-Murillo-Schlages (GMS) [D. O. Gericke *et al.*, Phys. Rev. E **65**, 036418 (2002)] based on a *T*-matrix method and the approach by Brown-Preston-Singleton [L. S. Brown *et al.*, Phys. Rep. **410**, 237 (2005)] agrees with the simulation data to within the error bars of the simulation. For smaller Coulomb logarithms, the GMS model is consistent with the simulation results. Landau-Spitzer models are consistent with the simulation data for $\mathcal{L} > 4$.

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I. INTRODUCTION

The strong temperature dependence of thermonuclear reaction rates suggests that even small deviations from equilibrium can yield differences in burn rates. Thus, the pursuit of ignition in the laboratory will benefit from accurate models of relaxation processes in hot dense plasmas. One of the greatest uncertainties in the nonequilibrium energy balance is the electron-ion temperature relaxation rate. Although there have been indirect measurements for cool dense matter [1], there is no experimental data in the regime of interest. However, this picture is changing rapidly, as electron-proton temperature equilibration data is forthcoming from the OMEGA laser at the University of Rochester. In addition, there are experiments planned at the National Ignition Facility and the ultrashort pulse laser facilities to measure electron-ion coupling rates in hot dense plasmas. Since large experiments are proposed and developed using complex radiation-hydrodynamic codes that contain simple formulas for energy transfer in the nonequilibrium plasma, any insight into the accuracy and realms of validity of the energy transfer equations used will have an impact on experimental design and analysis of on-going and future experiments. Theoretically, the challenge has been toward the development of temperature equilibration formulas for plasmas undergoing Coulomb collisions which are free from the divergences which plagued classical and semiclassical treatments of the problem. Here we take a complementary approach to hot dense plasmas by using molecular dynamics (MD) techniques. We use this method to test recent theoretical models and compare with standard results.

The electron-proton coupling rate was first calculated by Landau [2] and Spitzer (LS) [3] for classical plasmas with weak collisions. They write the electron-proton temperature exchange rate ($1/\tau_{pe}$) in the form

$$\frac{1}{\tau_{pe}} = \frac{8\sqrt{2}\pi n_p Z^2 e^4}{3m_e m_p c^3} \left\{ \frac{k_B T_e}{m_e c^2} + \frac{k_B T_p}{m_p c^2} \right\}^{-3/2} \mathcal{L} \equiv \frac{\mathcal{L}}{J_{LS}}, \quad (1)$$

where J_{LS} is the LS prefactor, n_e (n_p) are the electron (ion) number densities, $Z=1$ is the proton charge, T_e (T_p) are the

electron (ion) temperatures, and k_B is the Boltzmann constant. \mathcal{L} is the Coulomb logarithm containing details of the collision process. LS used

$$\mathcal{L}_{LS} = \ln(b_{max}/b_{min}), \quad (2)$$

where b_{max} and b_{min} are impact parameter cutoffs needed to remove divergences that arose from their treatment. b_{min} is chosen to be a minimum impact parameter consistent with plasma conditions, such as the classical distance of closest approach ($b_C = Ze^2/k_B T$). At high temperatures, b_{min} is often modified to include quantum diffraction effects by introducing the length scale of the electron thermal deBroglie wavelength $\Lambda = \sqrt{2\pi\hbar^2/m_e k_B T_e}$. Typically b_{max} is chosen to be a screening length arising from collective plasma phenomena, such as the Debye length $\lambda_D = \sqrt{k_B T_e / 4\pi e^2 n_e}$.

The presence of *ad hoc* cutoffs and other inconsistencies led researchers to derive kinetic equations without cutoffs [4–7]. The essence of these theories is the inclusion of strong scattering in the presence of dynamical collective (screening) behavior. Two such theories have recently been proposed: Gericke, Murillo, and Schlages (GMS) [7] and Brown, Preston and Singleton (BPS) [8]. GMS applied these ideas to dense plasma temperature equilibration. They investigated various approximations in the evaluation of \mathcal{L} , including issues with trajectories and cutoffs, and provided four different evaluations of the relaxation rate based on quantum kinetic theory. From their numerical work, GMS suggested an effective Coulomb logarithm [9]

$$\mathcal{L}_{GMS} = \frac{1}{2} \ln(1 + [\lambda_D^2 + R_{ion}^2] / [\Lambda^2 / 8\pi + b_C^2]), \quad (3)$$

where $R_{ion} = (3/4\pi n_p)^{1/3}$ is the ion sphere radius. This expression was described by GMS as the best fit to their full *T*-matrix theory.

BPS and Brown and Singleton [10] used dimensional continuation to obtain expressions for the electron-ion coupling rate accurate to second order in the plasma coupling param-

eter. The method is applicable to both degenerate and nondegenerate electrons. For the nondegenerate case, they derive

$$\mathcal{L}_{BPS} = \log(\lambda_D/\Lambda) + [\log(16\pi) - \gamma - 1]/2, \quad (4)$$

where γ is the Euler constant.

The most direct method of studying temperature equilibration in the classical limit is with numerical simulation; strong, collective scattering at all length scales is the forte of MD. Hansen and McDonald (HM) [11] explored temperature equilibration in dense hydrogen using MD, comparing their results against a LS model with $\mathcal{L}_{LS} = \ln(2\pi\lambda_D/\Lambda)$. However, the HM simulations involved a very small number of particles ($N=128$) with presumably large error bars. Here, we expand upon their calculations to not only reassess the HM result, but also compare with the modern approaches of GMS and BPS.

II. MOLECULAR DYNAMICS: SIMULATIONS AND RESULTS

MD simulations are applied to two-temperature systems of charged particles in a cubic cell with periodic boundary conditions. The MD is performed with a fully parallel code using a basic leapfrog method [12] with the Coulomb interaction evaluated by an Ewald summation [13,14]. Because the classical Coulomb many-body problem is unstable for attractive interactions, we employ semiclassical potentials that reduce the Coulomb interaction on short length scales in order to prevent unphysical deeply-bound states. We tested several forms of the diffractive [16,17] and Pauli [18,19] terms for these potentials. The resulting equilibration times typically vary by less than 15%, which is within the statistical error of the MD data. The similarity is not unexpected, since most semiclassical potentials resemble one another above 10 eV [21]. We report results using the semiclassical potential in HM [11],

$$V_{ab}(r) = \frac{Z_a Z_b e^2}{r} [1 - \exp(-2\pi r/\Lambda_{ab})] + k_B T \ln 2 \times \exp(-4\pi r^2/\Lambda_{ab}^2/\ln 2) \delta_{ae} \delta_{be}, \quad (5)$$

where $\Lambda_{ab} = \sqrt{2\pi\hbar^2/\mu_{ab}k_B T}$, μ_{ab} is the reduced mass, and $T = T_e$ except when a and b are both protons when $T = T_p$. The potentials as written are temperature (and hence time) dependent. For simulations run to full temperature equilibration, we allow the temperature-dependent parameters to evolve with time using a smoothed exponential average of the instantaneous MD value. For shorter runs (during which the temperature did not vary much) we held these parameters fixed.

Simulations were run long enough to extract a relaxation time (typically 10% of τ), with some strong-coupling cases continued to complete equilibration. We obtain τ_{pe} by fitting the temperature over a brief interval,

$$\frac{dT_e}{dt} = \frac{T_p - T_e}{\tau_{ep}}, \quad \frac{dT_p}{dt} = \frac{T_e - T_p}{\tau_{pe}}. \quad (6)$$

We choose the time step to conserve total energy over the entire simulation ($\Delta E/E < 10^{-4}$) when using fixed potentials.

Typically, Δt ranges from 5×10^{-5} to 10^{-3} fs. Any drift in the energy is tightly controlled, as artificial heating can distort the true relaxation rate. In long runs, the potentials change slowly as the temperature relaxes. Although energy is not conserved in these cases, the total energy change remains less than 3%. In practice, τ_{pe} calculated from the time-dependent potential is within 10–15 % of the result for the constant potential.

Convergence with respect to system size is tested by employing various particle numbers N ranging from $N=128$ (the number that HM employed), to as many as $N=64\,000$. The results reported here use $N=1024$. Statistical uncertainty for each case is estimated by computing the relaxation rate from equivalent samples (from 8 to 64) of a microcanonical ensemble and then taking the average and standard deviation.

The nonequilibrium system is prepared using two separate Langevin thermostats for protons and electrons. Initial configurations are sampled from a stationary distribution obtained after 10^5 – 10^7 time steps. The thermostats are then removed, and the species are allowed to undergo (microcanonical) collisional relaxation for approximately 10^6 time steps. Sensitivity to initial conditions is studied by sampling multiple independent systems from a microcanonical ensemble and/or by discarding a portion of the initial temperature evolution.

The basic relaxation process is collisional energy exchange, which must be related to temperature relaxation through an equation of state. We define temperature in the MD simulations directly from the mean-squared velocities, and so no such relation is needed. However, ideal gas equations of state were assumed in the GMS, BPS, and LS models with which we compare. Previous studies of equation of state corrections [15] suggest that the ideal gas equation state is accurate in the regime that we are considering. Although the MD temperature relaxation is asymmetric in the strong-coupling cases, we find $|dT_e/dt|$ and $|dT_p/dt|$ differ by only about 10%. Thus, we only report $1/\tau^* = 1/\tau_{pe} + 1/\tau_{ep} \approx 2/\tau_{pe}$.

Table I lists the set of initial conditions for 15 different systems. The ensemble average temperature relaxation, τ^* (calculated from $d\Delta T/dt = \Delta T/\tau^*$), and the standard deviation, σ , are in femtoseconds. A range of initial conditions were chosen to span the weakly to strongly coupled and the degenerate to nondegenerate regimes. We include two sets of initial conditions considered by HM (cases *J* and *K*). In most cases, hydrogen plasma is simulated using the true electron-proton mass ratio of 1:1836. In case *L*, the cold electrons were replaced with cold protons in order to shorten the required simulation time. Cases *M*_{1–3} involve a comparison of electron-proton and positron-proton systems and will be discussed below. Cases *F* and *G* have degenerate electrons. Degeneracy effects are treated in neither the classical MD simulations nor in the LS, GMS, or BPS models. Hence, the models can be directly compared to the simulations even for those cases when comparisons with experiment would be questionable.

III. COMPARISON WITH THEORY

Figure 1 shows MD results for case *K* run to near-full relaxation using potentials that are implicitly time dependent

TABLE I. Density, initial electron, and ion temperature, relaxation time, and standard deviation of the MD simulations.

Case	n_i (cm ⁻³)	T_e (eV)	T_i (eV)	τ^* (fs)	σ (fs)
A	10 ²⁰	10.0	20.0	2.04 × 10 ⁴	4.9 × 10 ³
B	10 ²⁰	30.0	60.0	7.89 × 10 ⁴	4.3 × 10 ⁴
C	10 ²²	10.0	20.0	5.23 × 10 ²	1.7 × 10 ²
D	10 ²²	30.0	60.0	1.73 × 10 ³	6.6 × 10 ²
E	10 ²²	100.0	200.0	6.45 × 10 ³	2.2 × 10 ³
F	10 ²⁴	10.0	20.0	8.87 × 10 ¹	3.5 × 10 ¹
G	10 ²⁴	30.0	60.0	8.27 × 10 ¹	3.3 × 10 ¹
H	10 ²⁴	100.0	200.0	1.72 × 10 ²	6.2 × 10 ¹
I	10 ²⁴	300.0	600.0	4.17 × 10 ²	8.0 × 10 ¹
J	1.61 × 10 ²⁴	29.9	80.1	20.2 × 10 ¹	5.3
K	1.61 × 10 ²⁴	91.47	12.1	1.20 × 10 ²	1.7 × 10 ¹
L	10 ²⁰	100.0	200.0	3.65 × 10 ⁵	3.2 × 10 ⁵
M ₁	10 ²⁰	10.0	40.0	2.05 × 10 ⁴	3.0 × 10 ³
M ₂	10 ²⁰	10.0	40.0	2.18 × 10 ⁴	4.5 × 10 ³
M ₃	10 ²⁰	10.0	40.0	2.28 × 10 ⁴	9.6 × 10 ³

(temperature dependent). We also display predictions for LS, GMS, and BPS. The MD data in Fig. 1 is most closely matched by BPS (although this is partly fortuitous, as will be seen below) followed by GMS. The LS model predicts the fastest relaxation, exceeding MD by about a factor of 2. This disagreement contradicts the conclusion reached by HM. At the same time, our τ^* for cases *J* and *K* agree with those reported by HM. We attribute the discrepancy to incon-

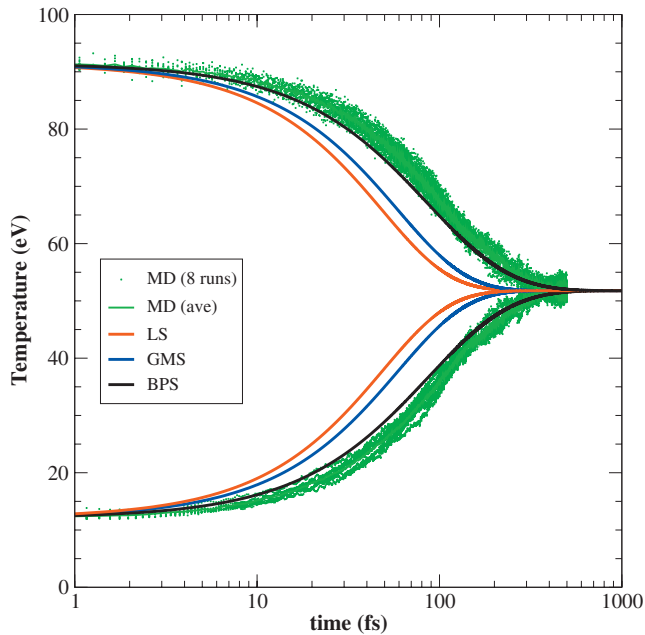


FIG. 1. (Color) Electron (top curves) and proton (bottom curves) temperature relaxation is shown based on MD, GMS, LS, and BPS for case *K*. The MD results are shown by points from several simulations, with a line through the average. Note that all approaches relax slower than LS.

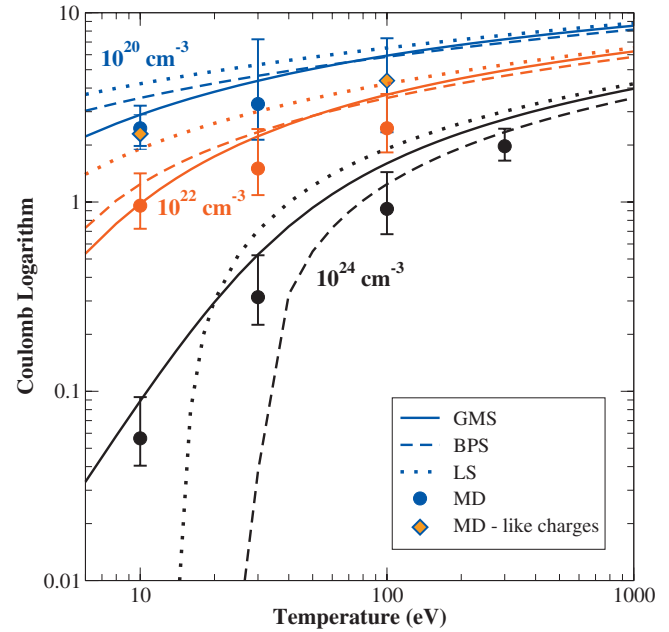


FIG. 2. (Color) Theoretical (GMS [solid], BPS [dashed], and LS [dotted]) and MD calculations of \mathcal{L} as a function of initial T_e for densities 10²⁰ cm⁻³, 10²² cm⁻³, and 10²⁴ cm⁻³ (blue, red, and black, respectively). Diamond symbols indicate results obtained from like-charge MD simulations. Additional detail is in the text.

sistent definitions of τ_{pe} , τ_{LS} , and τ^* : τ_{LS} is properly equal to τ_{pe} , which is $2\tau^*$ (not τ^*) if $\tau_{pe} \equiv \tau_{ep}$. As previously noted by HM, however, ambiguities in the b_{min} and b_{max} may be sufficient to accommodate this difference.

To make comparisons of our MD results with theoretical predictions more transparent, we define an effective Coulomb logarithm as $\mathcal{L}_{MD} \equiv 2J_{LS}/\tau^*$. This result is then compared with the theoretical prediction for \mathcal{L} coming from LS, GMS, and BPS. Figure 2 shows simulation results for \mathcal{L}_{MD} with error bars along with theoretical predictions for \mathcal{L}_{GMS} (solid), \mathcal{L}_{BPS} (dashed), and \mathcal{L}_{LS} (dotted) as a function of initial electron temperature. Numerical results and analytic expressions for \mathcal{L} are arranged according to density; $n = 10^{20}, 10^{22}$, and 10^{24} cm⁻³ (blue, red, and black, respectively).

In regions where it is expected to be applicable, we find that LS systematically overestimates the effective Coulomb logarithm and thus predicts a relaxation rate that is too fast relative to the MD results. For plasmas with $\mathcal{L} > 1$, the MD results are consistent with both the GMS and BPS, suggesting that approaches beyond LS are indeed more predictive. As expected, BPS increasingly underestimates the relaxation rate for $\mathcal{L} < 1$; BPS is not intended for use in this regime. For the case shown in Fig. 1, the underestimation at lower temperatures compensates for an overestimation at early times, making agreement with this simulation fortuitously good. As is evident from Fig. 2, this would not be the case in general [22]. We find that GMS captures the qualitative variation of \mathcal{L} over a surprisingly broad range of density and temperature. Further discrimination between these theories in the region where they are expected to be most accurate (low density and high temperature) is not possible given the large

uncertainties present in our current MD simulations. However, our results suggest that validation of these theories could be accomplished with carefully controlled experiments [23] and larger (and longer) simulations that further reduce statistical error.

Finally, LS predicts identical equilibration rates for like-charge and opposite charge systems. We tested this by performing three sets of simulations at the same density and temperatures (case M_{1-3} in Table I.) We simulated electrons-protons (M_1) and positrons-protons (M_2 , plotted in Fig. 2) using Eq. (6), and positrons-protons using a pure $1/r$ Coulomb potential (M_3). The relaxation rates for all three cases agree to within our error bars, suggesting that energy transfer in these systems is occurring predominately on length scales longer than the thermal deBroglie wavelength.

IV. CONCLUSIONS

We have performed MD simulations of the temperature relaxation process in hot dense hydrogen. We investigated systems containing as many large as 64 000 particles, finding that $N \approx 1000$ particles is sufficient for most cases we considered. Our simulations span a large range of temperature and density parameter space, including the first simulations in the low-density high-temperature limit. The main processes in the molecular dynamics are treated exactly: screen-

ing, collective modes, and collisions. We have compared two different forms of the semiclassical potential and the pure $1/r$ form and have found only very slight differences; as such, we believe that our results are not sensitive to the choice of the semiclassical form of the potential for the range of conditions considered.

For the weakly coupled plasmas where $\mathcal{L} \gtrsim 1$, the simulations are consistent with both GMS and BPS. In contrast, the LS approach systematically overestimates the relaxation rate. In the limit of high temperature and low density, all models are in agreement, however. Our MD results show that LS is accurate for $\mathcal{L} > 4$, rather than the usual restriction of $\mathcal{L} > 10$, in agreement with previous work [7,20]. More modern approaches exemplified here by GMS and BPS clearly extend the accessible parameter space closer to $\mathcal{L} \sim 1$, with GMS providing a reasonable description of the MD data even for $\mathcal{L} < 1$.

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- [1] P. Celliers, A. Ng, G. Xu, and A. Forsman, *Phys. Rev. Lett.* **68**, 2305 (1992); A. Ng, P. Celliers, G. Xu, and A. Forsman, *Phys. Rev. E* **52**, 4299 (1995).
 - [2] L. D. Landau, *Phys. Z. Sowjetunion* **10**, 154 (1936); *Zh. Eksp. Teor. Fiz.* **7**, 203 (1937).
 - [3] L. Spitzer, Jr., *Physics of Fully Ionized Gases*, 2nd ed. (Interscience, New York, 1962).
 - [4] E. A. Frieman and D. C. Book, *Phys. Fluids* **6**, 1700 (1963).
 - [5] H. A. Gould and H. E. DeWitt, *Phys. Rev.* **155**, 68 (1967).
 - [6] M. W. C. Dharma-wardana and F. Perrot, *Phys. Rev. E* **58**, 3705 (1998).
 - [7] D. O. Gericke, M. S. Murillo, and M. Schlanges, *Phys. Rev. E* **65**, 036418 (2002).
 - [8] L. S. Brown, D. L. Preston, and R. L. Singleton, Jr., *Phys. Rep.* **410**, 237 (2005).
 - [9] This is model 6 in Ref. [7]. Note that we have recast Eqs. (3) and (4) to use a consistent definition of the thermal deBroglie wavelength.
 - [10] L. S. Brown and R. L. Singleton, Jr., *Phys. Rev. E* **76**, 066404 (2007).
 - [11] J. P. Hansen and I. R. McDonald, *Phys. Lett.* **97A**, 42 (1983).
 - [12] F. H. Streitz, J. N. Glosli, and M. V. Patel, *Phys. Rev. Lett.* **96**, 225701 (2006).
 - [13] P. Ewald, *Ann. Phys.* **64**, 253 (1921).
 - [14] S. W. DeLeeuw, J. W. Perram, and E. R. Smith, *Proc. R. Soc. London, Ser. A* **373**, 27 (1980).
 - [15] D. O. Gericke and M. S. Murillo, in *Proceedings on Inertial Fusion Sciences and Applications 2003* (ANS, La Grange Park, IL, 2004), p. 1002.
 - [16] C. S. Jones and M. S. Murillo, *High Energy Density Phys.* **3**, 379 (2007).
 - [17] T. Dunn and A. A. Broyles, *Phys. Rev.* **157**, 156 (1967).
 - [18] H. Minoo, M. M. Gombert, and C. Deutsch, *Phys. Rev. A* **23**, 924 (1981).
 - [19] G. Kelbg, *Ann. Phys.* **12**, 219 (1963).
 - [20] C.-K. Li and R. D. Petrasso, *Phys. Rev. Lett.* **70**, 3059 (1993).
 - [21] A. V. Filinov, V. O. Golubnychiy, M. Bonitz, W. Ebeling, and J. W. Dufty, *Phys. Rev. E* **70**, 046411 (2004).
 - [22] For densities near 10^{24} cm^{-3} , \mathcal{L}_{BPS} happens to cross \mathcal{L}_{MD} between 50 and 100 eV, midrange of the relaxation in Fig. 1.
 - [23] J. M. Taccetti, R. P. Shurter, J. P. Roberts, J. F. Benage, B. Graden, B. Haberle, M. S. Murillo, B. Vigil, and F. J. Wysocki, *J. Phys. A* **39**, 4347 (2006).