Molecular Dynamics Simulations of Electron-Ion Temperature Equilibration in an SF_6 Plasma

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We use classical molecular dynamics to investigate electron-ion temperature equilibration in a two-temperature SF_6 plasma. We choose a density of $1.0 \times 10^{19} SF_6$ molecules per cm^3 and initial temperatures of $T_e = 100$ eV and $T_S = T_F = 15$ eV, in accordance with experiments currently underway at Los Alamos National Laboratory. Our computed relaxation time lies between two oft-used variants of the Landau-Spitzer relaxation formula which invoke static screening. Discrepancies are also found when comparing to the predictions made by more recent theoretical approaches. These differences should be large enough to be measured in the upcoming experiments.

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The problem of electron-ion temperature equilibration in a plasma has enjoyed renewed interest in the last two decades, partly as a result of experimental work on cold laser-produced plasmas [1], and partly due to interest in the pursuit of fusion ignition in the laboratory [2]. This nonequilibrium process, in which there is a slow relaxation to a common temperature in a system with unequal electron and ion temperatures, is crucial for understanding the overall energy balance in a fusion-burning plasma, where the burn rate is strongly temperature dependent. Theoretical approaches based on kinetic theory, such as those involving the Boltzmann equation, have been applied to nonequilibrium problems of this type. However, complications arise when dealing with systems whose particles interact via the Coulomb interaction. In these systems for which the collision integrals are typically divergent, special methods have been developed [3,4], yet definitive answers for temperature equilibration rates are still lacking, as candidate approaches can produce markedly different predictions.

The problem of temperature equilibration was first addressed by Landau [5] and Spitzer [6] (LS), who used the Fokker-Planck equation to derive an equilibration rate for electron temperature T_e, given an ion temperature T_i,

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

where $m_{e,i}$ are the electron and ion masses, $n_{e,i}$ are the number densities, and Z_i is the ion charge. The so-called Coulomb logarithm, $\ln\lambda_{ie}$, is a quantity arising from the (assumed 2-body) collision integral, usually written as the ratio of maximum to minimum impact parameters as $\ln(b_{\max}/b_{\min})$. Its appearance is a consequence of the divergence of the Coulomb cross section when integrated over impact parameter, and its determination is troublesome in a many-body Coulomb system where screening and quantum diffraction effects play a role. In applications of LS, $b_{\max}$ is taken to be a Debye screening length involving either (a) only electrons (LS_e) λ_{De}, or (b) electrons and ions (LS_{tot}) λ_{Dtot}, while $b_{\min}$ is chosen to be (a) the classical distance of closest approach, $b_0 = Z_i e^2/k_B T_e$ (neglecting center-of-mass effects), or (b) the electron thermal de Broglie wavelength $\Lambda = \hbar\sqrt{2\pi/m_e k_B T_e}$. This prescription is expected to be valid in the limit of large $\ln\lambda_{ie}$.

The two choices for $b_{\max}$ mentioned above (λ_{De} and λ_{Dtot}) are both suspect, as they arise from static screening approximations to a dynamical problem. In a general treatment [7,8], the equilibration rate $1/\tau_{ie}$ involves a frequency integral over the Fourier transform of the screened electron-ion interaction, $v(\mathbf{k})/\epsilon(\mathbf{k}, \omega)$, where v is the Coulomb interaction and ϵ is the plasma dielectric function. LS_e and LS_{tot} assume that the frequencies most important for the computation of $1/\tau_{ie}$ are well below the electron plasma frequency (and even below the ion plasma frequency for LS_{tot}). Though this is partially true for some plasmas, since the heavier ions respond more slowly than the lighter electrons, it is not strictly true in general.

Recently, there have been several theoretical and computational works which aim to study temperature equilibration. Theoretical works include the contributions by Dharma-wardana and Perrot (DP) [8], Gericke, Murillo and Schlanges (GMS) [9], and Brown, Preston, and Singleton (BPS) [10]. DP used a many-body Green function approach and considered dynamical screening (including coupled electronic and ionic modes) and partial degeneracy. Their results suggested marked differences from LS and were not of the general form of Eq. (1). GMS used a T-matrix approach which addressed strong electron-ion coupling but with a partial neglect of dynami-

cal screening [9]. Their results could be represented by a form similar to Eq. (1), but with a $b_{\max}$ interpolating between Λ_{De} and the ion sphere radius, and a $b_{\min}$ interpolating between b_0 and Λ. BPS strictly addressed only weak plasma coupling and used dimensional continuation to provide a justification of LS-type expressions but without *ad hoc* cutoffs. Their analytic result for $1/\tau_{ie}$ reduces to the form of Eq. (1) with an *effective* $\ln\lambda_{ie}$ involving the ratio of λ_{De} and Λ. This is appropriate for plasmas with $\Lambda > b_0$ where quantum diffractive effects dominate the closest encounters. The recent computational studies have all been done with classical molecular dynamics (MD) and have focused on temperature equilibration in a multi-eV hydrogen plasma [11–13]. The results of these studies showed that deviations from LS's approach are to be expected, even for moderate values of $\ln\lambda_{ie}$, with one contribution [11] demonstrating that the GMS model works surprisingly well for small effective $\ln\lambda_{ie}$.

To date, there have been no experimental studies that have determined $1/\tau_{ie}$ well enough to test these various predictions. An experiment to do just this is currently underway at Los Alamos National Laboratory, in which a gas jet of SF_6 molecules is excited with a laser and both electron and ion temperatures will be independently measured in the approach to equilibrium [14]. The choice of molecule, as well as conditions of density and initial temperatures have been made to optimize the quality of the electron and ion temperature measurements. Densities in the jet are expected to be highly uniform and of order $1.0 \times 10^{19} SF_6/cm^3$. T_e and T_i are expected to be roughly 100–150 eV and 10–20 eV, respectively. Though the ionization stages will vary with time, modeling has shown that the dominant processes will produce a plasma with $Z_S = 11$ and $Z_F = 7$ (leading to 53 free electrons per SF_6) for much of the equilibration [14]. These plasma conditions are characterized by λ_{De} (≈ 32 Å) greater than the ion sphere radius ($\approx$16 Å for F), λ_{Dtot} (≈ 6 Å) *less than* the ion sphere radius, and a b_0 ($\approx$1 Å for e-F) slightly greater than Λ ($\approx$0.7 Å). The large difference between λ_{De} and λ_{Dtot} is due to the sizable Z and the low T_i for this plasma. This leads to $\ln\lambda_{\mathrm{LS}_e} \approx 3$ and $\ln\lambda_{\mathrm{LStot}} \approx 1$, putting the plasma into a regime in which LS's approach is beginning to be suspect [15], yet the electron-ion coupling parameter, $\Gamma_{ie} \approx 0.15$, is still rather small. However, these conditions also produce a strong ion-ion coupling, $\Gamma_{ii} \approx 3$. This is a regime not yet studied with MD. In addition, the fact that $b_0 > \Lambda$ suggests that this plasma would be especially amenable to treatment using classical MD.

We present nonequilibrium classical MD simulations for temperature equilibration for this idealization of an SF_6 plasma ($Z_S = 11$, $Z_F = 7$) at a density and set of initial temperatures relevant for this experiment. It is shown that the MD results produce a relaxation notably slower than that of BPS and LS_e, and faster than that of $\mathrm{LS}_{\mathrm{tot}}$. We expect that these differences are large enough to be observable in the planned experiment.

MD simulations were performed with the ddcMD code, which uses a velocity Verlet integration with a particle centric domain decomposition method [16]. Long-range Coulomb forces were calculated by the particle-particle-particle mesh method [17]. Since the classical Coulomb many-body problem is unstable for attractive interactions, we employed semiclassical potentials derived for electron and ion systems at temperature T [18,19], in which the short-range part of the Coulomb interaction is softened within a distance set by Λ. Our earlier studies for a hydrogen plasma also employed these potentials and demonstrated an insensitivity to the specific choice of their form for the cases studied [11].

We placed 1000 S, 6000 F, and 53000 electrons (corresponding to 1000 dissociated SF_6 molecules at a density of 1.0×10^{19} cm^{-3}) in a box with periodic boundary conditions. Initial coordinates were chosen randomly and then adjusted with a Monte Carlo procedure that ensured that no two particles were too close to each other. The S^{+11} and F^{+7} ions were modeled as point particles with no internal degrees of freedom. To establish the initial temperature separation, electron and ion Langevin thermostats with target temperatures $T_e \approx 100$ eV and $T_i \approx 15$ eV were applied for 10^5–10^7 time steps, after which the thermostat was removed and the system was allowed to evolve microcanonically. We chose the time step to conserve total energy over the duration of the simulation to within $\Delta E/E < 10^{-4}$, which then resulted in $\Delta t \approx 5 \times 10^{-5}$ fs.

Given that the mass ratio between ions and electrons is very large ($\frac{m_S}{m_e} \sim 6 \times 10^4$), accurate simulations would require extremely long simulation times to produce significant electron-ion temperature relaxation. To reduce this time we performed a sequence of temperature equilibration simulations with ion masses which are a fraction of their physical masses, $m_i \rightarrow \alpha m_i$, and then extrapolated to the physical masses ($\alpha = 1$) by dividing the simulation time by α [20]. This would be correct if the only dependence of the equilibration rate on ion mass was that appearing in the *first* factor in Eq. (1) (i.e., $1/\tau_{ie} \propto 1/m_i$). However, (a) the factor in brackets depends weakly on m_i, (b) $\ln\lambda_{ie}$ may depend weakly on m_i through reduced-mass effects in the distance of closest approach, and (c) the true equilibration rate may not strictly be of the form given by Eq. (1). Regarding (a) and (b), we note that for LS-type theories, the mass-scaling we outline here is extremely accurate as long as $\alpha > 0.0001$. Regarding (c), we show below that for a range of α between 0.0005 and 0.01, our scaled MD results exhibit only minor variations; we discuss the subtleties of these variations below.

Figure 1 shows the electron and ion temperatures as a function of time. The value $\alpha = 0.0005$, together with the appropriate scaled time, was used to obtain the MD results shown as thick symbols. Thin lines represent various theoretical results discussed below. The final equilibrated temperature ≈ 90 eV is much closer to the initial electron temperature because the number of free electrons is

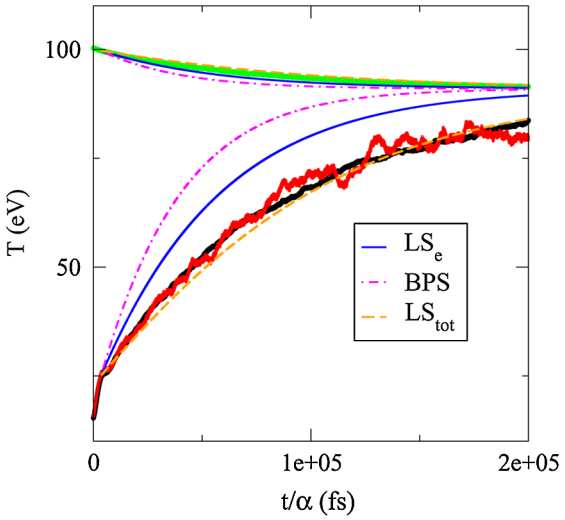

FIG. 1 (color online). MD results for electron [green (light gray) solid line] and ion [S (red [medium gray] solid line), F (black solid line)] temperatures as a function of scaled time ($t \rightarrow t/\alpha$) for $\alpha = 0.0005$, compared with rate-equation results using LS_e (LS with only electron screening; blue [dark gray] solid line), BPS (magenta [dash-dotted line]), and LS_{tot} (LS using the total Debye length; orange [dashed line]). Initial conditions for LS_e, LS_{tot}, and BPS were set at $t = 3634$ fs), at which the ion temperatures are ≈ 25 eV.

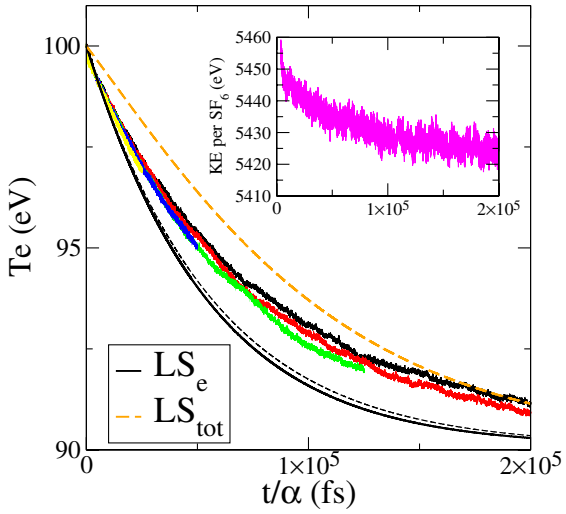

FIG. 2 (color online). Detailed view of the electron temperature relaxation, together with the LS_e and LS_{tot} predictions. Different colors represent the different ion mass scalings: $\alpha = 0.0005$ (black), 0.001 (red [medium gray]), 0.002 (green [light gray]), 0.005 (blue [dark gray]), 0.01 (yellow [lightest gray]), in which the time axis has been scaled according to $t \rightarrow t/\alpha$. The LS_e (LS with only electron screening) result for $\alpha = 0.0005$ is represented by the thin grey line just above the ($\alpha = 1$) LS_e curve. LS_{tot} (LS using the total Debye length) is shown as the orange dashed line. Inset shows the total (electron + ion) kinetic energy as a function of time for $\alpha = 0.0005$.

much larger than the number of ions, resulting in a larger heat capacity for the electron subsystem. This also gives rise to larger fluctuations in the ion temperature during the simulation, as is evidenced by the increased noise in the T_i versus t curves. Note also that the S and F temperatures remain close throughout the simulation. This is expected, due to the larger energy transfer between near equal-mass species as compared to that for very unequal masses. Figure 2 shows a detailed view of the electron temperature relaxation. Here, we have shifted the MD data so that $T_e = 100$ eV at $t = 0$ (the actual starting temperatures were slightly different, owing to fluctuations during the application of the thermostat). The five curves represent runs for different values of mass scaling, α. Note that the variations between the curves are small compared to the difference between any one of them and the LS predictions, also shown and discussed below.

In order to compare the MD results to those of the available theories for electron-ion temperature relaxation, we assume that the time-evolution of the temperatures (T_e, T_S, T_F) are governed by the coupled rate equations: $dT_e/dt = (T_S - T_e)/\tau_{eS} + (T_F - T_e)/\tau_{eF}$ (along with others for dT_S/dt and dT_F/dt, obtained by permuting the indices). We also assume that the total energy of the system is equal to its total kinetic energy, thereby neglecting deviations of the specific heats of the subsystems from their ideal gas values; such deviations are discussed below. Conservation of total kinetic energy, $n_e dT_e/dt + n_S dT_S/dt + n_F dT_F/dt = 0$, then requires that $\tau_{Se} = (n_S/n_e)\tau_{eS}$, etc. These reciprocity relations are obeyed by the Spitzer-like formula for τ given above since it was derived from a Fokker-Planck prescription which makes the ideal gas specific heat assumption. The values of τ_{eS} and τ_{eF} are taken from Eq. (1) assuming various forms for $\ln\lambda_{ie}$. Generalizing Eq. (1) to apply to ion-ion equilibration, the LS theory predicts a negative $\ln\lambda_{SF}$ in these conditions due to strong ion-ion coupling. Knowing that the kinetics of ion-ion equilibration is much faster than electron-ion, we circumvent this problem by setting $\ln\lambda_{SF} = 2$, independent of temperatures. Our results for electron-ion equilibration are completely insensitive to this choice.

The thin lines of Fig. 1 show the rate-equation predictions for the time evolution of the temperatures for various choices of $\ln\lambda_{ie}$. Note that T_S is essentially equal to T_F for these predictions. LS_e predicts significantly faster electron-ion equilibration than that shown by the MD data. BPS predicts a still faster equilibration. This can be understood by examining the form of $\ln\lambda_{BPS}$: the *effective* b_{min} for BPS is Λ; since b_0 is greater than Λ for these plasma conditions, $\ln\lambda_{BPS}$ is greater than $\ln\lambda_{LS_e}$, and BPS relaxes even faster than LS_e. Since the intended application of BPS was to more weakly coupled plasmas [10], the discrepancy here comes as no surprise. GMS (not shown) also predicts a faster relaxation than MD, and is almost identical to LS_e.

In addition to these comparisons, we see that even though our effective $\ln\lambda$ is as large as 3, the LS_e approach is not accurate.

The fact that the MD data exhibits a notably slower relaxation than LS_e suggests that one or a few of the assumptions inherent in the LS_e theory are inapplicable. One such assumption is static screening, in which the $b_{\max}$ is taken to be λ_{De}. The thin curve labeled LS_{tot} in Figs. 1 and 2 represents the result of LS with a different static screening assumption: $b_{\max} = \lambda_{Dtot}$. This produces a relaxation significantly slower than LS_e, a result of the large reduction of the screening length due to the highly-charged ions. The MD results lie between the two, suggesting that a correct accounting of dynamical screening may lead to better agreement. In addition, since λ_{Dtot} is less than the ion sphere radius, *a strong-screening approach going beyond a Debye picture* would likely be needed to correctly account for ion screening here. Other assumptions of LS, such as the complete neglect of (classical, in this case) bound states, and the consideration of only 2-body scattering, may also be culprits. Nevertheless, we note that the variations between the T_e curves for different ion masses shown in Fig. 2 are larger than that predicted from the more detailed scaling implied by the LS formula in Eq. (1); LS_e for $\alpha = 0.0005$ is shown as the grey curve just above the $a = 1$ LS_e curve in Fig. 2. These differences are in the right direction (slower relaxation for more equal ion and electron masses) to suggest that dynamical screening may be important, since the choice of equal ion and electron masses would clearly favor the LS_{tot} approach [21].

Careful study of Figs. 1 and 2 reveals an asymmetry: For early times, T_i is closer to LS_{tot} while T_e is farther from LS_{tot}. This means that the total kinetic energy is not strictly conserved, as assumed in the standard rate-equation treatment; the inset to Fig. 2 shows the total kinetic energy decreasing with time. This is the result of total energy conservation and the ions being strongly coupled. Since the ions begin cold, the (screened) ion subsystem has a lower *potential* energy at the start of the run than at the end, when the heated ions are less correlated. The magnitude of this kinetic energy decrease is consistent with the predicted potential energy increase for a model Yukawa system with parameters chosen to match this SF_6 plasma [22].

In summary, we presented MD results for electron-ion temperature equilibration in an SF_6 plasma. For the case of $n_{SF_6} = 1.0 \times 10^{19}/\mathrm{cm}^3$ and initial temperatures $T_e = 100$ eV and $T_{S,F} = 15$–25 eV, the relaxation is significantly slower than that of the Landau-Spitzer formula with electron-only screening, and is faster than that of Landau-Spitzer with a Debye screening length including both electrons and ions. Other candidate models, such as that of BPS, do no better. We remind the reader that the study presented here is for fixed, time-independent S and F ionizations. The true time-dependent ionization levels present in the experiment may alter the picture somewhat, though we expect the gross features to be similar to those presented here. Our MD results suggest that deviations from these different models are large enough to be seen in the upcoming SF_6 experiment.

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