Quantum corrections and bound-state effects in the energy relaxation of hot dense Hydrogen.

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Simple analytic formulae for energy relaxation (ER) in electron-ion systems, with quantum corrections, ion dynamics and RPA-type screening are presented. ER in the presence of bound electrons is examined in view of recent simulations for ER in hydrogen in the range $10^{20}-10^{24}$ electrons/cc.

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Introduction – The problem of energy relaxation (ER) in hot electron-ion systems arises in astrophysics, fusion physics, condensed-matter physics and chemistry\cite{1,2}. The large difference in masses $m_e, m_i$ between electrons and ions simplifies the problem somewhat since quasi-equilibrium systems, with an electron temperature $T_e$, and an ion temperature $T_i$ can occur. Such two-temperature plasmas exist in material systems as diverse as warm dense matter \cite{3}, ultra-cold plasmas \cite{4}, hot semiconductors \cite{5}, and dense deuterium \cite{6}. The strong temperature dependence of thermonuclear processes imply that estimated burn rates depend sensitively on the accuracy of the temperature relaxation (TR) theory used. Thus a number of recent studies\cite{1,7,8} have examined various aspects of two-temperature dense hydrogen and ER in regimes of densities and temperatures relevant to inertial confinement fusion (ICF)\cite{6}.

The earliest theories of ER in plasmas are due to Landau \cite{10} and Spitzer \cite{11} (denoted L-S). The L-S approach is applicable to weakly interacting fully-ionized plasmas in the classical regime. It is the Rutherford Coulomb scattering formula applied to Maxwellian distributions of ions and electrons. Analyses using the Fermi golden-rule (FGR) and coupled-mode (CM) extensions were given by Dharma-wardana et al. \cite{1,12,13,14}. Fokker-Plank approaches\cite{12}, kinetic-equation methods based on the relaxation of the one-particle distributions\cite{15,16}, methods based on expansions in the coupling constant\cite{17} or related techniques\cite{18}, have been explored in recent studies\cite{17}. Molecular dynamics (MD) was used by Hansen and McDonald, (denoted HM)\cite{19}, using classical potentials which incorporate the cutoffs used in the Coulomb logarithm of L-S theory. The recent studies of Refs. \cite{1,8} report more careful applications of the HM approach. The FGR and CM approaches have been experimentally tested in semiconductor plasmas, but no experimental results of TR are as yet available for hot dense plasmas.

In fully-ionized hydrogen plasma the particle charges $Z_p, Z_e$ are $\pm 1$. The mean electron- and proton densities $n$ and $\rho$ are identical. The ratio of a typical Coulomb energy to the kinetic energy becomes, in the classical regime, $\Gamma = 1/(r_s T)$, where $T$ is the temperature in energy units, and $r_s = [3/(4\pi n)]^{1/3}$ is the radius of the Wigner-Seitz sphere of an electron or a proton. The properties of partially degenerate plasmas require two independent parameters, e.g., both $\Gamma$ and $\theta = T/E_F$, where $E_F = (3^{2/3} \pi n)^{2/3}/2$ is the Fermi energy. Thus the regime of densities and temperatures studied in Ref. \cite{8} involve, at one extreme, $r_s = 25.25, \theta = 127$ at $T_e=10$ eV, for $n_e = 10^{20}$ electrons/cc, while the another extreme is $r_s = 1.172, \theta = 0.274$, where $n_e = 10^{24}$ electrons/cc at $T_e=10$ eV. The latter is a significantly degenerate plasma where the validity of classical-simulation methods is suspect. The system at $r_s \sim 25, n_e = 10^{20}$ electrons/cc at $T_e$ 10 eV contains 1s to 3d bound states, but the system is essentially ionized since the bound-state occupations are negligible. On the other hand, the plasma at $n_e = 10^{22}$ electrons/cc, $T_e = 10$ eV, i.e., $r_s=5.441$ is $\sim 82\%$ ionized at $T_e = 10$ eV and carries a 1s bound state at an energy of -0.458 a.u., when calculated using the atom-plasma codes implemented by Perrot et al\cite{20}. The effect of such bound states cannot be included in the classical simulation method of Ref. \cite{3}.

In this communication we use the FGR approach and derive a simple analytic formula inclusive of leading quantum corrections, screening and ion-dynamics for for TR in the fully ionized limit, and compare it with the results from recent classical simulations \cite{8}. We then consider the effect of bound states, since they can have a significant effect on energy relaxation. Many complex processes become possible, but the system with a single hydrogenic boundstate is a useful case for sharpening our understanding of this relatively unknown regime where no previous results are available.

Quantum transition rates.– Assuming that $T_e > T_i$ to be specific, ER occurs via energy transfer from the excited modes of the electron sub-system to the cold modes in the ion subsystem. The spectrum of the modes of the species $j$ is given by the spectral function $A_j(q, \omega, T_j)$. These spectral functions are given by the imaginary parts of the corresponding dynamic response functions $\chi^j(\vec{k}, \omega)$, e.g, Eq. (16) of Ref. \cite{14}. The ER rate evaluated within the Fermi golden rule, $R_{fgr}$ can be expressed in terms of the response functions of the plasma as given

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in Eqs. (4)-(7) of Ref.[13], and Eq. (15) of Ref. [14]:

$$R_{fr} = \frac{\delta E}{\delta t} = \int \frac{d^3k}{(2\pi)^3} \frac{\omega d\omega}{2\pi} (\Delta B) F_{ep}$$

$$\Delta B = \coth(\omega/2T_e) - \coth(\omega/2T_p)$$

$$F_{ep} = \left| (V_{ep}(k))^2 \right| \Delta \left[ \chi^p(\vec{k}, \omega) \right] \Delta \left[ \chi^c(\vec{k}, \omega) \right]$$

In the above $\delta E/\delta t$ is the rate of change of the energy of the system, for time steps $\delta t$ significantly greater than the equilibrations times $\tau_p, \tau_p$ which establish $T_e$ and $T_p$ of each subsystem. The relaxation of the whole system is determined by $\tau_{ep}$ such that $\tau_{ep} >> \tau_p > \tau_e$. For brevity we write $\delta E/\delta t$ as $dE/dt$. We have used the spherical symmetry of the plasma to write scalars $q, k$ instead of $\vec{q}, \vec{k}$ to simplify the notation. The non-interacting response function $\chi_0(q, \omega, T)$ at arbitrary degeneracies was given by Khanna and Gylde[21], and are used here in the generalized RPA $\chi^j(q, \omega, T)$ form, $j = e, p$ where, for example, temperature-dependent local field corrections $G_{ee}(k)$ may be included[22]. If $T_e, T_i$ are both sufficiently large that $\Delta B \rightarrow 2(T_e - T_i)/\omega$, and if the electron chemical potential $\mu_e \leq 0$, useful analytical approximations become available. The possibility of extracting a temperature-relaxation time $\tau_{ep}$ from the relaxation rate exists only in this regime. Neglecting interactions, $E$ becomes the kinetic energy. Using non-interacting classical forms for $\Im \chi_0(k, \omega)$ in Eq. 1 we obtain the well known Landau-Spitzer (L-S) form for the temperature relaxation time $\tau$, viz.,

$$1/\tau = \frac{2}{3n} \omega_p^2 \omega_{pp}^2 \left| (2\pi T_{ep})/m_{ep} \right|^{-3/2} \mathcal{L}$$

$$\mathcal{L} = \log(k_{max}/k_{min})$$

$$T_{ep}/m_{ep} = T_e/m_e + T_p/M_P, \ \omega_{pp}^2 = 4\pi n/m_j$$

Here $\omega_p$ is the plasma frequency of the species $j = e, p$. The effective temperature and the effective mass of the colliding pair are $T_{ep}$ and $m_{ep}$, with $T_j$ in energy units. $\mathcal{L}$ is the “Coulomb logarithm”. It depends on $k_{min}$ and $k_{max}$ i.e., momentum cutoffs (or impact parameters) used for modeling the unscreened Coulomb collision. If interacting response functions (e.g, RPA) are used, single-particle modes become replaced almost completely by plasmon modes, and the interactions become dynamically screened. Any type of “static screening” must satisfy the $f$-sum rule if true results are to be avoided. The safest procedure is to do the $\omega$-integration imposing the $f$-sum rule[14]. Then ion dynamics are automatically preserved, and Eq. 1 simplifies to:

$$\frac{1}{\tau} = \frac{2}{3n} \omega_p^2 \omega_{pp}^2 \int_0^{\infty} \frac{2}{\pi} \left[ \frac{\partial}{\partial \omega} \Im \chi^{ee}(k, \omega) \right]_{\omega=0} dk$$

We approximate $\Im \partial \chi^{ee}/\partial \omega |_{\omega=0}$ as:

$$\Im \partial \chi^{ee}/\partial \omega |_{\omega=0} = \frac{\Im \partial \chi^{ee}/\partial \omega |_{\omega=0}}{\{1 + k_{sc}^2/k^2\}^2}$$

FIG. 1: The Landau-Spitzer Coulomb Logarithm ($\mathcal{L}$) is compared with the classical MD simulations[8] and the analytic Coulomb factor $\mathcal{Q}$, Eq. 15 derived from the FGR result.

The anti-symmetry of the imaginary part has been used in the above. The electron-screening wavevector $k_{sc}$ at any degeneracy is obtained from the small-$q$ limit of the finite-$T$ Lindhard function. The $k \rightarrow 0$-local field correction, $G_{00}^c$ at arbitrary degeneracy[22] can also be included in $k_{sc}$ via the following definitions.

$$(k_{sc})^2 = \frac{2}{\pi} (2T_e)^{1/2} I_{-1/2}(\mu_e/T_e)$$

$$I_\nu(x) = \int_0^\infty \frac{dy y^\nu e^{-y}}{e^y - x + 1}, \ \nu \geq 1/2$$

$$k_{sc} = k_{sc}^0 \left[ 1 - (\epsilon_{ee}/\epsilon_0)^{1/2} \right]$$

However, in this study we find it sufficient to use he Debye-Hükel form $k_{sc} = k_{D_H}$ for electron screening. In approximating $\Im \partial \chi^{ee}/\partial \omega$ we retain terms up to second order in $\hbar$ as displayed explicitly below:

$$\Im \chi_0^{ee} = -\frac{\pi}{2T_e} \frac{2\omega_p^2}{\pi k} e^{-\omega_p^2/4\hbar \omega/2T_e} \sinh(\hbar \omega/2T_e)$$

Then Eq. 7 can be reduced to the form:

$$1/\tau = \frac{2}{3n} \omega_p^2 \omega_{pp}^2 \left| (2\pi T_{ep}/m_{ep}) \right|^{-3/2} \mathcal{Q}$$

$$\mathcal{Q} = \frac{1}{2} \left[ e^\mu E_i(p_e)(p_e + 1) - 1 \right]$$
\[ p_e = \frac{k^2_{B} T_e}{8 T_e} \]  
\[ Ei(x) = \int_{x}^{\infty} \frac{e^{-t} dt}{t} \]  
\( (15) \)  
\( (16) \)

The exponential integral \( Ei(x) \) of Eq. 16 is evaluated numerically via standard subroutines. Thus we see that the “Coulomb factor” \( Q \) is exactly analogous to the “Coulomb logarithm” of Eq. 6 but without \( ad~hoc \) cutoffs. \( Q \) contains leading-order quantum corrections, ion-dynamics and electron screening. The expression for \( Q \) should be compared with a similar expression given by Brown et al[17].

\[ \mathcal{L}_{bps} = \tfrac{1}{2} \left[ \log(1/p_e) - \gamma - 1 \right] \]  
\( (17) \)

where \( \gamma = 0.5772 \) is the Euler constant. At high \( T_e \), this result approaches the L-S form more rapidly than \( Q \).

The interactions between the ion- and electron modes lead to ion-acoustic modes (coupled modes) [1]. The H-plasmas treated here may be considered relatively weekly coupled plasmas, and the correction from coupled-mode effects will be neglected. The results shown in Fig. 1 suggest that the classical potentials etc., used for \( r_s = 5.44 \) and \( r_s = 25.25 \) need reconsideration, since these are weakly coupled plasmas where FGR methods should give good agreement. As seen from Fig. 2 the numerical results from \( Q \) and \( \mathcal{L}_{bps} \) are very similar. The case \( r_s \approx 1 \) has been discussed in more detail elsewhere [1].

Energy Relaxation in the presence of bound states—The nuclei in the H-plasma with 10\(^{22}\) electrons/cc, \( r_s = 5.44 \) carry a single 1s bound state of energy \( \sim 0.46 \) a.u. with the effective charge \( Z \) varying from 0.82 at \( T_e = 10 \) eV, to \( \sim 1.0 \) at \( T_e > 200 \) eV. If we use an “average ion” picture, we have a gas of ions with change \( Z \), a boundstate occupation \( n_{1s} = 1 - Z \) electrons per ion, and the remaining (“free”) electrons distributed in continuum states. The electrons in the bound states have an effective mass \( m_{e,ps} \approx m_e = 1 \), and hence equilibrate rapidly with the hot electrons in the continuum. That is, the distribution \( n_{1s} \), and hence the degree of ionization \( Z \) is determined by the hot electron temperature \( T_e \). (See fig. 2 for a plot of (1 – \( Z \)). The center of mass (CM) motion of the ions and bound electrons is determined by the kinetic energy of the mass \( M_{CM} = M_p + m_e \approx M_p \), i.e., CM motion is at the temperature \( T_p \). The energy exchanging collisions are between the hot electrons and the ions. The bound electrons (in their 1s states) do not interact with their own binding nuclei. The energy mismatch in the spectral functions ensures that they do not significantly interact with other ions carrying bound states. This “average-ion” picture holds if the temperature relaxation time \( \tau \) is sufficiently long compared to the ionization-equilibration time \( \tau_Z \). This picture is applicable to the system discussed in this study, and the only source of energy relaxation is via Coulomb collisions.

An alternative picture holds if \( \tau \) is comparable to \( \tau_Z \). Then the plasma consists of nonionized atomic hydrogen with fully occupied 1s states at some temperature \( T_a \) of the atoms, free electrons at \( T_e \) and protons at \( T_p \). The atom temperature \( T_a \) has to be self consistently determined via a theory of ionizing collisions as well as the Coulomb collisions discussed so far. This regime occurs for low \( T_e \), \( T_p \), and is not considered here.

The energy \( E_s \) of the electron subsystem, and the energy \( E_i \) of the CM-ion subsystem in the average-ion model with hydrogenic states \( \epsilon_{\nu} \), occupations \( n_{\nu} \) (\( \nu = n, l, m \) being the set of quantum numbers of the state) is given by:

\[ E_i = 3nT_p/2 + E_{int} \]  
\[ E_e = 3nZT_e/2 + n(1 - Z) \sum_{\nu}(\epsilon_{\nu} - \mu_{a})n_{\nu} \]  
\[ \approx 3nZT_e/2 + n(1 - Z)^2 \epsilon_{1s} + E_{e, int} \]  
\( (18) \)  
\( (19) \)  
\( (20) \)

The terms \( E_{int} \) involve contributions from particle interactions, embedding effects, continuum lowering effects etc., which have been discussed at length in Ref. [24]. These effects can be neglected for the weekly-coupled H-plasmas studied here. The last equation limits the problem to one bound state. The effects arising from the chemical potential \( \mu_a = \mu_e(T_e) + \mu_p(T_p) + \delta \mu_a \) will be neglected in all the species, free and bound, as they are
found to be small. Then the time dependence $dE_j/dt$ can be replace by $dE_j/(dT_j)(dT_j/dt)$ and we attempt to construct a formula for temperature relaxation. Unlike in simple classical plasmas, it is no longer possible to define a temperature relaxation time $\tau$. Hence, to be specific we may thermostat $T_i$ and consider the relaxation rate $dT_e/dt$, with $(Z \neq 1)$ and without $(Z = 1)$ bound-state effects. Thus we have:

$$dT_e/dt|_{Z=1} = (T_e - T_i)C_{pe}Q(p_e)$$  \hspace{1cm} (21)

$$C_{pe} = \frac{2}{3n}a_p^2\omega_p^2\{2\pi T_{pe}/m_{pe}\}^{-3/2}$$ \hspace{1cm} (22)

$$dT_e/dt|_{Z\neq1} = (T_e - T_i)\frac{Z^2C_{pe}}{B_{pe}}Q(Zp_e)$$ \hspace{1cm} (23)

$$\frac{1}{B_{pe}} = \left[ Z + \frac{4}{3}(1 - Z)\epsilon_{1s} \right] \frac{dZ}{dT_e}$$

This result shows that the effective ionization $Z$ enters approximately as $Z^2$ in the prefactor. The dependence of the ionization temperature, viz., $dT_e/dT_e$ was obtained from a DFT calculation\[20\]. It may be argued that some of these corrections are more part of the specific heat of the electron system inclusive of bound states, and do not belong to the collision dynamics. However, merely to compare the $Z = 1, Z \neq 1$ systems, we define

$$Q_b = \{Z^3/B_{pe}\}Q(Zp_e)$$ \hspace{1cm} (24)

We display $Q$ and $Q_b$ using the Eqs.\[21\] in Fig.\[2\] together with the classical MD, $L_{bps}$ and L-S results which treat only the fully ionized state. The inset to the figure shows the degree of association (1 $-$ $Z$) as a function of temperature, calculated using density-functional methods\[20\].

**Conclusion.** We presented analytic formulae with quantum corrections, ion dynamics and screening. They have been applied to weakly coupled H-plasmas ($r_s = 5.44, 25.25$) where the physical approximations are felt to be reliable. The differences between the classical MD simulations and our results suggest the need for further review of the classical simulations as well as the physical theory. The effect of the bound states on a quantity nominally similar to the Coulomb logarithm has also been presented for the H-plasma at the density $10^{22}$ e/cc. This too is an area which requires further study and poses a novel challenge to simulations..

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