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Stochastic substitute for coupled rate equations in the modeling of highly ionized transient plasmas

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Plasmas produced by intense laser pulses incident on solid targets often do not satisfy the conditions for local thermodynamic equilibrium, and so cannot be modeled by transport equations relying on equations of state. A proper description involves an excessively large number of coupled rate equations connecting many quantum states of numerous species having different degrees of ionization. Here we pursue a recent suggestion to model the plasma by a few dominant states perturbed by a stochastic driving force. The driving force is taken to be a Poisson impulse process, giving a Langevin equation which is equivalent to a Fokker-Planck equation for the probability density governing the distribution of electron density. An approximate solution to the Langevin equation permits calculation of the characteristic relaxation rate. An exact stationary solution to the Fokker-Planck equation is given as a function of the strength of the stochastic driving force. This stationary solution is used, along with a Laplace transform, to convert the Fokker-Planck equation to one of Schrödinger type. We consider using the classical Hamiltonian formalism and the WKB method to obtain the time-dependent solution.

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The interaction of a high-irradiance laser with a solid target creates several spatial regions characterized by low-density plasma in the outer parts (called corona) and high-density plasma in the inner domain close to the solid target. In most cases, local thermodynamic equilibrium (LTE) is not reached in the corona.

In LTE the distribution of state densities, both in ground or excited states, is given by the Saha and Boltzmann equations. More generally, all laws of thermodynamic equilibrium are valid for electrons and ions. However, LTE is satisfied mainly for high-density plasmas where the collisions between electrons themselves produce an equilibrium. For LTE to happen it is necessary that collisional interactions be much more frequent than the corresponding dissipative radiative processes. For example [1], such a condition for an aluminum plasma with an electron temperature of about 1 keV requires electron densities larger than 10^{23} cm^{-3} in order to achieve LTE for the He-like ground state and H-like first excited state. Taking into account that a corona is defined for electron densities $n_e < n_c \simeq 10^{21}/\lambda^2$ [cm^{-3}], where n_c is the critical density [1] and λ is the laser wavelength in micrometers, it is evident that LTE is not achieved in a "hot" dilute plasma.

With the advent of short laser pulses [2], a non-steady-state plasma is created. Moreover, for these short pulses ($\sim$psec) even the electrons might be far from LTE so that an electron temperature is not properly defined.

In an LTE regime the equations of state (EOS) are well defined [3]. The EOS relations are necessary in order to solve the plasma fluid equations. It is evident that in a plasma where LTE is not satisfied, such as a laser-produced corona or a picosecond laser-produced plasma, one cannot define a temperature and therefore EOS relations do not exist. In this case one has to solve a set of rate equations describing the changes of the densities of the different possible quantum states.

The general kinetic rate equations for each type of ion with a density N_i are with A_{ji} being the total rate for the transition from level j to i and B_{ij} the transition rate from i to any j state:

$$\frac{dN_i}{dt} = \sum_j (A_{ji}N_j - B_{ij}N_i) , \quad i=1,\ldots,\nu . \tag{1}$$

Both A_{ji} and B_{ij} contain collisional and radiative terms. The numerical solutions of these kinetic equations are treated by a large number of computer codes that have been developed to simulate non-LTE experiments [4].

In this paper we follow the recently suggested idea [5] of describing a non-LTE plasma by a few ion states in interaction with electrons sustained in a medium of "noise." The noise describes the extremely large number of states not considered explicitly as well as all the collision terms not taken into account.

We will follow a scheme with three species of ion coupled through ionization (I) and recombination rates (R), neglecting both excitation and deexcitation. N_{j-i}, N_j, and N_{j+1} are the number densities for these three ion species. It is also assumed that rates are due only to collisions with free electrons, avoiding the three-body recombination that works near the LTE region. Also, radiation terms are neglected. Then, the kinetic equations for the relative population $n_j = N_j/N_t$, with N_t being the total number density, are the following:

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$$\frac{dn_{j-1}}{dt}=n_eR_jn_j-n_eI_{j-1}n_{j-1}\ , \tag{2}$$

$$\frac{dn_j}{dt}=n_eI_{j-1}n_{j-1}+n_eR_{j+1}n_{j+1}-n_e(I_j+R_j)n_j\ , \tag{3}$$

with the closure equation

$$1=n_{j-1}+n_j+n_{j+1} \tag{4}$$

and the constraint equation for charge conservation

$$x\equiv\frac{n_e}{N_t}=(j-1)n_{j-1}+jn_j+(j+1)n_{j+1}\ . \tag{5}$$

The values of R_j (radiative recombination rate) and I_j (collisional ionization rate) are given in the literature in Refs. [5–8].

Equations (2)–(5) are four equations with four dimensionless unknowns: n_{j-1}, n_j, n_{j+1}, and x. An algebraic combination of these equations yields

$$\frac{dx}{dt}=Ax^2+Bx\ , \tag{6}$$

where we have assumed $(d^2/dt^2)(\ln x)\simeq 0$. A and B are given by

$$A=-\frac{N_t(R_{j+1}R_j+R_{j+1}I_{j-1}+I_{j-1}I_j)}{R_{j+1}+R_j+I_j+I_{j-1}} \tag{7}$$

and

$$B=\frac{N_t[(j+1)I_jI_{j-1}+jR_{j+1}I_{j-1}+(j-1)R_jR_{j+1}]}{R_{j+1}+R_j+I_j+I_{j-1}}\ . \tag{8}$$

From Eqs. (7) and (8) one gets $A<0$ and $B>0$. The potential ϕ describing the nonlinear damped anharmonic oscillator of equation (6) is given by

$$\phi(x)=-\frac{A}{3}x^3-\frac{B}{2}x^2\ , \tag{9}$$

where the equation of motion in the electron density space is

$$\frac{dx}{dt}=-\frac{\partial\phi}{\partial x}\ . \tag{10}$$

The potential ϕ has a minimum at

$$x=-\frac{B}{A}=\frac{(j+1)I_jI_{j-1}+jI_{j-1}R_{j+1}+(j-1)R_jR_{j+1}}{R_{j+1}R_j+R_{j+1}I_{j-1}+I_{j-1}I_j}>0\ . \tag{11}$$

The equation of "motion" (6) is of the Bernoulle type and has the solution

$$x(t)=\left[\left[\frac{1}{x_0}+\frac{A}{B}\right]e^{-Bt}-\frac{A}{B}\right]^{-1}\ , \tag{12}$$

where x_0 is the initial condition $[x(t=0)]$. For $t\to\infty$, or more practically $t\gg t_R$, where t_R is the relaxation time given by $1/B$, one gets the steady-state solution given by (11).

Let us consider a lithiumlike iron plasma ($Z=26$, $j=23$), with an electron energy distribution about 600 eV and $\rho=0.002\ \text{g cm}^{-3}$ (i.e., $n_i=2.1\times10^{19}\ \text{cm}^{-3}$). For $E_{23}=850$ eV and $E_{24}=-1699$ eV, obtained with a hydrogenic atomic model [9], one gets

$$R_{23}=1.565\times10^{-12}\ \text{cm}^3\,\text{s}^{-1}\ ,$$
$$R_{24}=1.633\times10^{-12}\ \text{cm}^3\,\text{s}^{-1}\ , \tag{13}$$
$$I_{22}=I_{23}=1.075\times10^{-12}\ \text{cm}^3\,\text{s}^{-1}\ .$$

From Eqs. (7) and (8),

$$A=-2.29\times10^{7}\,\text{s}^{-1}\ ,\quad B=5.22\times10^{8}\,\text{s}^{-1}\ , \tag{14}$$

and using Eq. (11) the average ionization is obtained: $x=22.8$. Then the relaxation time for the numerical example is $t_R=B^{-1}\simeq2\times10^{-9}$ s. The relaxation time scales with electron plasma density n_e as

$$t_R=(2\ \text{ns})\left[\frac{2.1\times10^{19}\ \text{cm}^{-3}}{n_e}\right]x\ . \tag{15}$$

For example, for a critical plasma density of 9×10^{21} cm^{-3} [i.e., for 3ω laser frequency obtained from a Nd:YAG (neodymium-doped yttrium aluminum garnet)] one gets a value of 100 ps for the relaxation time.

It is suggested that the collision terms, radiative and all other interaction sources not taken explicitly into account in Eq. (6) be described by a "noise term." In particular, Eq. (6) is changed into a Langevin equation

$$\frac{dx}{dt}=Ax^2+Bx+\sqrt{g}\,W\ , \tag{16}$$

where the noise $\sqrt{g}\,W$ is a random "force" in density space

$$\sqrt{g}\,W(t)=\sqrt{g}\sum_j(-1)^{\epsilon_j}\delta(t-t_j)\ , \tag{17}$$

g being the size of the random impulse, $\varepsilon_j=0$ or 1 is a random variable, and t_j a random time sequence. The random fluctuation W satisfies

$$\langle W(t)\rangle=0\ ;\ \langle W(t)W(t')\rangle=\delta(t-t)\ , \tag{18}$$

where $\langle\ \rangle$ means a statistical average over the direction of the "impulses" ε_j and the time t_j. Equation (16) can be written via the potential ϕ,

$$\frac{dx}{dt}=-\frac{\partial\phi}{\partial x}+\sqrt{g}\,W\ , \tag{19}$$

where $-\partial\phi/\partial x$ is a deterministic "force" and $\sqrt{g}\,W(t)$ a random "force." The stochastic driving force ought to have two parameters, namely, the pulse intensity (g) and the rate at which the pulses occur. However, for a large number of states not taken into account directly, but assumed to contribute to the noise, one gets on single parameter: the rate at which the pulses occur is absorbed into a combined strength parameter.

The Langevin equation (19) is equivalent to the Fokker-Planck equation [10–12]

$$\frac{df}{dt}=\frac{\partial}{\partial x}\left[\frac{\partial\phi}{\partial x}f\right]+\tfrac{1}{2}g\frac{\partial^2 f}{\partial x^2}\ , \tag{20}$$

where $f(x,t)$ is the probability density for the variable x. This equation is assumed to be equivalent to the full set of rate equations. The diffusion term, the second term on the right-hand side of Eq. (20) in density space, describes the extremely large number of states not taken explicitly into account. Experimentally we expect to measure an average density $\langle x\rangle$ given by

$$\langle x\rangle(g,t)=\int f(x,t)x\,dx\ , \tag{21}$$

so that we have a one-parameter theory described by g. The stationary state, $df/dt=0$, is given from (21) as

$$f(x)=f_0e^{-2\phi/g}\ , \tag{22}$$

f_0 being a renormalization factor. At equilibrium the distribution f is maximal for a minimum potential ϕ.

The Langevin equation (19) leads to the Fokker-Planck equation (20) for the probability density $f(x)$. By substituting into (20)

$$f(x,t)=e^{-\phi(x)/g}\Psi_\lambda(x)e^{-\lambda t/g}\ , \tag{23}$$

one obtains a Schrödinger equation for $\Psi_\lambda(x)$,

$$H\Psi_\lambda(x)=\lambda\Psi_\lambda(x)\ , \tag{24}$$

where the Hamiltonian H is given by

$$H=-\frac{1}{2}g^2\frac{\partial^2}{\partial x^2}+\Phi \tag{25}$$

and the new potential Φ is related to the old potential by

$$\Phi=\frac{1}{2}\left[\frac{\partial\phi}{\partial x}\right]^2-\frac{1}{2}g\frac{\partial^2\phi}{\partial x^2}$$
$$=\frac{1}{2}(A^2x^4+2ABx^3+B^2x^2)+gAx+\frac{g}{2}B\ . \tag{26}$$

The constant $\frac{1}{2}gB$ can be discarded since Φ is defined up to a constant. This potential has an extremum for $\partial\Phi/\partial x=0$, implying

$$x^3+\frac{3}{2}\frac{A}{B}x^2+\frac{1}{2}\frac{B^2}{A^2}x+\frac{g}{2A}=0\ . \tag{27}$$

In analyzing the roots of Eq. (27) one defines

$$D=-\frac{1}{1728}\frac{B^6}{A^6}+\frac{g^2}{16A^2}\ , \tag{28}$$

so that for $D>0$ one has one real and two complex roots, for $D=0$ one has three real roots where one of these is a double root, while for $D<0$ one gets three real distinctive roots. For the $D=0$ case one gets from Eq. (28)

$$g\equiv g_0=\frac{1}{\sqrt{108}}\frac{B^3}{A^2}=0.096\frac{B^3}{A^2}\ \mathrm{s}^{-1}\ . \tag{29}$$

Substituting the values of A and B [Eq. (14)] for the Li-like ion plasma, one obtains

$$\tau_0\equiv\frac{1}{g_0}=38.45\ \mathrm{ps}\ , \tag{30}$$

which defines a time scale for the "noise." For a "large" noise $g>g_0$ the potential has one minimum, while for a "small" noise $g<g_0$ the potential [Eq. (26)] Φ has two minima and one maximum (see Fig. 1).

For $g=0$ the potential Φ has a minimum at $x=-B/A$, the same as that of ϕ [Eq. (9)]. For a "small" noise, i.e., $g\ll g_0$, one can find a minimum near $-B/A$. Using perturbation theory one gets for the minimum of Φ the value

$$\hat{x}=-\frac{B}{A}\left[1-\frac{A^2}{B^3}g\right]\ , \tag{31}$$

where x has the physical meaning of equilibrium ionization. Since $B>0$ and $A<0$ the "shift" due to "noise" in the equilibrium ionization is positive. So noise increases the equilibrium average ionization.

The use of the WKB approximation in order to find the solution of the Schrödinger equation (24) yields

$$\Psi_\lambda=\sum_j c_j\left[\frac{ds_j}{dx}\right]^{-1/2}\exp\left[\frac{i}{g}s_j(x,\lambda)\right]\ , \tag{32}$$

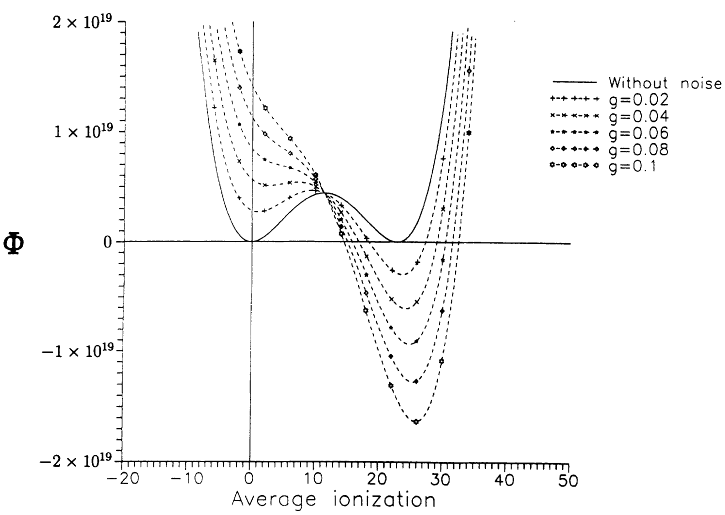

FIG. 1. Potential Φ (arbitrary units) versus average ionization (x) for several noise parameters (g). g is given in units of B^3/A^2.

where s_j are the solutions of the Hamilton-Jacobi equation

$$\frac{1}{2}\left[\frac{ds_j}{dx}\right]^2+\Phi=\lambda\ , \tag{33}$$

which satisfies

$$s_j(x,\lambda)=\int^x p_j dx\ . \tag{34}$$

The integration in (34) is done on the trajectories of Hamilton's equation

$$\frac{dp}{dt}=-\frac{\partial H}{\partial x}\ ,\quad \frac{dx}{dt}=\frac{\partial H}{\partial p}\ , \tag{35}$$

$$H=\tfrac{1}{2}p^2+\Phi(x)\ , \tag{36}$$

where Φ is given in Eq. (26).

In this paper the non-LTE plasmas are discussed. In these plasmas the temperature is not defined and therefore the thermodynamic equilibrium is not achieved. In particular, the equations of state are not well defined and therefore the "hydrodynamic" approach to these plasmas is not useful. For non-LTE plasmas one has to use transport equations (such as Boltzmann or Fokker-Planck) with source terms. The source terms are described by rate equations for the different species of the plasma. An ion species is defined by its ionization and quantum state. The plasma rate equations can be presented as a damped anharmonic oscillator in the density space. For this equation a potential ϕ is defined [see Eq. (9)] which describes the time development of the ionization in the plasma.

In a "complex" plasma, e.g., a laser-induced corona from a high-Z target, the number of ions species is extremely large so that a very large ($\rightarrow\infty$) number of rate equations must be considered. This approach is very difficult to follow and may be prohibitive on present available computers. Therefore, we suggest describing a non-LTE plasma by a few ion states in interaction with electrons sustained in a medium of "noise." The noise simulates the states not considered explicitly as well as the collision terms not taken into account. This noise is assumed to modify the damped anharmonic oscillator equation into a Langevin equation for the electron density. This Langevin equation is equivalent to the Fokker-Planck equation. A transformation is made to obtain a Schrödinger equation so that the set of the original rate equations is described by a Schrödinger equation with a potential Φ [Eqs. (24) and (25)]. This potential includes the "noise" so that we have a one-parameter (g) Hamiltonian which should describe the time development of the densities through Eq. (23).

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