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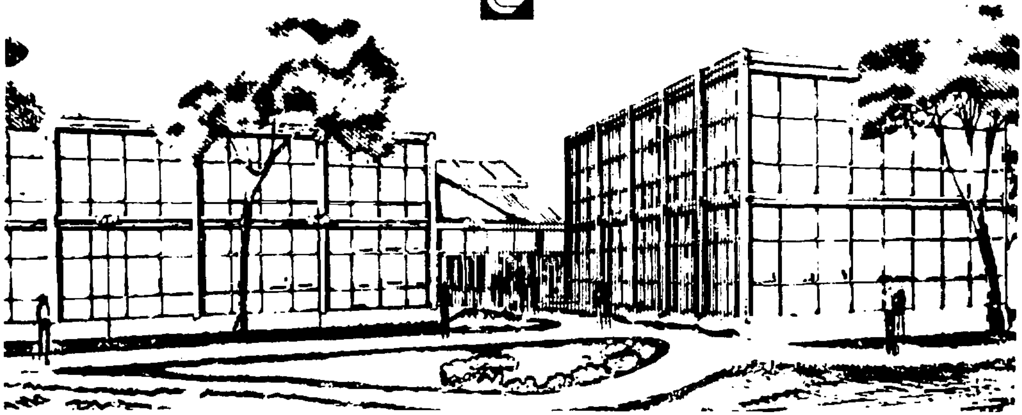

MASTER

Pressure Ionization in Laser-Fusion Target Simulation*

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ABSTRACT

Accurate simulation of high density target implosion requires material properties (ionization, pressure, energy, opacity and transport coefficients) at densities where bound electrons are significantly perturbed by neighboring atoms. In the LASNEX code, this data is supplied by tables and/or calculated from a Stromgren model for ionization equilibrium.

Improvements have been made in this model which aim at assuring thermodynamic consistency and obtaining better agreement with more elaborate calculations. Arbitrary degeneracy is allowed for the free electrons. Consistent Coulomb contributions to pressure and continuum lowering are obtained. A new pressure ionization scheme merges bound electrons into the continuum as a smooth function of density and the corresponding contribution to pressure is calculated. Results are shown for aluminum.

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Introduction

Accurate numerical simulation of laser-produced plasmas requires a variety of atomic properties of the target material including thermodynamic data (pressure and energy), transport coefficients (electrical and thermal conductivity) and optical absorption coefficients (group-mean opacities). Calculation of these quantities requires knowledge of the ionization state and atomic level populations. However, for the very dense plasmas produced by laser compression of solids, the well-known ionization models give poor results.[1-5]

The following problems are typical: (1) Complete recombination to the neutral atom at high densities, (2) large regions where the ionization $Z^*(V,T)$ is a constant corresponding to a closed-shell configuration, (3) abrupt jumps of Z^* with density, (4) poor convergence of iteration schemes, in which levels oscillate between being bound and unbound on successive iterations.[1-6] For high densities, the ionization-equilibrium model typically gives results which differ significantly from the TF or TFD theories. Although modest differences (10-25%) are expected from electron shell effects,[7] the ionization equilbrium (Saha Equation) calculations are as much as a factor of 2 off the TF or TFD results for high density and low temperature. In this range of temperature and density, there is good evidence that the TF theory is more accurate.[7]

This report describes an improved ionization-equilibrium model which deals with these problems in a practical way. The model is based on the existing LASNEX package for rate equations, equation of state and opacity[8,9] and the alterations described here are very tightly constrained by the need to avoid sweeping changes which would interfere with the many virtues of the existing code.

LASNEX solves hydrodynamic and transport equations using physical data from a variey of sources, including analytic formulas, subroutines and tabular data. EOS tables combining experimental data and the TFK (or TFD) theory have been used for LTE calculations while non-LTE problems require an inline EOS calculation. Improving the inline model so that it can be used for LTE calculation of compressed matter offers consistency and continuity between LTE and non-LTE methods. It also eliminates the need to produce and store EOS data for all possible mixtures of elements.

The changes described here yield credible results for plasmas at densities above normal solid densities. The EOS is thermodynamically consistent to high accuracy (better than 1%). The modified code still requires less than 10^{-3} sec per EOS point, so it is usable in-line. The changes have the incidental effect of improving the smoothness of the EOS at lower densities.

Average-Atom Model for Ions

The average-atom model discussed here was introduced by Stromgren in the 1920's. It was treated in great detail by Harris Mayer (1948)[10] and J. Green (1965).[11] In recent years it was developed as a rapid subroutine package by W. Lokke and collaborators at Lawrence

Livermore Laboratory.[8-9] We have begun from that subroutine, and modified the treatment of pressure ionization.

The subroutine used by LASNEX includes provision for a relativistic treatment based on the energy levels associated with the Dirac equation. In this report, the simpler non-relativistic scheme will be described. The formulas will refer to a single element, although the code also calculates mixtures of elements.

The model is based on a one-electron description of the atomic ionization state; the one-electron states of a self-consistent potential are approximately identified and each state is assigned a fractional occupation.

The full set of hydrogenic quantum numbers (n, l, m, σ) or (n, j, jz, σ) is truncated by assuming the energy depends only on the principle quantum number n. (This describes the statistical portion of the code; in the opacity calculation the orbital quantum numbers are also considered). The group of electrons having fixed n is referred to as a shell. Up to 10 shells are allowed.

The nth shell is characterized by the following quantities:

Z_n = screened charge seen by electrons of the nth shell.

ε_n = average energy of electrons in the nth shell $= -(Z_n/n^2) \times (13.6\ \text{eV})$.

r_n = average orbit radius for electrons in the nth shell $= (n^2/Z_n) \times (.529\ \text{Å})$.

D_n = shell degeneracy $= 2n^2\phi_n(\rho)$; $\phi_n(\rho)$ will be described

f_n = occupation = average number of electrons per state in the nth shell.

In terms of these quantities, the ionization state Z^* is

$$Z^* = Z - \sum_n f_n D_n$$

where Z is the nuclear charge. The population P_n (= number of electrons in the nth shell) is $f_n D_n$. The screened charge Z_n is obtained by an expression of the form

$$Z_n = Z - \sum_m \sigma_{nm} f_m D_m$$

where the summation includes the case $n = m$. The screening constants have been taken from H. Mayer (reference 10).

The novel feature of these equations is the expression for D_n, which contains a function $\phi_n(\rho)$ which serves to model the phenomenon of pressure ionization in an approximate way.

Pressure Ionization

The basic phenomenon of pressure ionization can be described very simply. The electrons of the nth shell have orbit radii which are approximately given by $r_n = (n^2/Z_n)(.529Å)$. When atoms are pushed together so that the average nuclear separation is less than $2 \times r_n$, the nth shell will (obviously) be severely distorted.

Accurate quantum calculations show that this distortion consists of the coupling of localized atomic states to form bands of extended wavefunctions which propagate between atoms. At high compressions, these bands blend into free-electron states.

Our model of these changes is simply to reduce the number of states of a given shell from $2n^2$ to zero as a smooth function of density ρ. The density function selected scales the reduction of D_n on the ratio $r_n{}^o/R_0$ where

$$R_0 = (3/4\pi N_i)^{1/3}$$

is the ion-sphere radius (N_i is the ion number density) and $r_n{}^o$ is the radius of the nth shell in the neutral free atom. The scaling function is tuned to produce reasonable results; we have examined the simple form

$$D_n = \frac{2n^2}{1 + \left(a \frac{r_n{}^o}{R_0}\right)^b}$$

which appears quite satisfactory.

Several comments will help to motivate this choice. First, it is intentional that D_n is calculated from r_n^o (the free atom orbit radius) rather than r_n (the orbit radius at the current density and temperature). The reason is that the subsequent thermodynamic calculations are tractable if D_n depends only on density (r_n has a slight dependence on the occupations f_n and hence on temperature). For the occupied levels, r_n and r_n^o are in fact not very different.

Second, the function shown for D_n turns a shell off over a range of densities. This is intentional, and simulates two real phenomena in pressure ionization. One is the tendency of the low-l (s-states and p-states) to be pressure-ionized before the other states of a given shell.[12] The other is the gradual nature of the relaxation of bands to the free-electron limit as density rises; this relaxation does not abruptly occur when a given band crosses zero energy.

C. A. Rouse has described a pressure-ionization scheme, based on a modified Saha equation, in which ionic states are switched out of existence by a multiplier keyed to the density.[2] Our model differs from his in two respects: First, we use the one-electron orbital picture instead of itemizing ions and their excited states; second, we have pursued the effects of the pressure-ionization to obtain consistent thermodynamic results.

Thermodynamic Consistency

A general requirement for a reasonable equation-of-state model is imposed by thermodynamic consistency. This requirement is not merely academic aesthetics; the U.S. Patent Office refuses to patent devices which appear to be perpetual motion machines of the first or second types.

The condition for thermodynamic consistency is simply derived fom the Helmholtz free energy $F(V,T)$, whose partial derivatives are

$$\text{pressure } P = -(\partial F/\partial V)_T$$

$$\text{entropy } S = -(\partial F/\partial T)_V$$

The energy is given by

$$\text{Energy } E = F + TS = F - T\,(\partial F/\partial T)_V$$

It follows directly that

$$T\left(\frac{\partial P}{\partial T}\right)_V = \left(\frac{\partial E}{\partial V}\right)_T + P$$

and this is the condition for thermodynamic consistency.

In a code such as LASNEX the temperature of a fluid is advanced according to

$$\left(\frac{\partial E}{\partial T}\right)_V \Delta T = -\Delta V\left[\left(\frac{\partial E}{\partial V}\right)_T + P\right] + \Delta U$$

where ΔU is the energy gained by transport and other nonhydrodynamic processes. Since $P\Delta V$ is the change in kinetic energy, this equation shows explicit energy conservation. For a plasma $\left(\frac{\partial E}{\partial V}\right)_T + P > 0$, but for very degenerate matter the two terms are nearly equal and opposite. The use of inconsistent expressions for P and E then can cause erroneous temperature changes. The use of $T\left(\frac{\partial P}{\partial T}\right)_V$ instead of $\left(\frac{\partial E}{\partial V}\right)_T + P$ in the temperature equation generally yields reasonable values for ΔT, but absolute energy conservation is lost unless the EOS is thermodynamically consistent.

Free Energy for Dense Plasma

The ionization model is easily forced to be thermodynamically consistent by deriving it from a free energy function. The appropriate expression proves to be

$$F_{total} = F_{ion} + F_{free} = \Delta F_{ion\text{-}sphere}$$

$$F_{ion} = E_{ion} - TS_{ion}$$

$$S_{ion} = -k \sum_n D_n \left[f_n \log f_n + (1 - f_n) \log (1 - f_n) \right]$$

$$E_{ion} = \sum_n \varepsilon_n f_n D_n$$

where the quantities (ε_n, f_n, D_n) have all been defined above. F_{free} is the free energy of a free electron gas (the complicated formula is available in the literature) of Z^* electrons per ion, where

$$Z^* = Z - \sum_n f_n D_n$$

$\Delta F_{ion\text{-}sphere}$ is the continuum-lowering, a function of Z^*, V_{ion}, T (V_{ion} is the volume per ion). In our work this is taken to be

$$\Delta F_{ion\ sphere} = -\frac{9}{10} \frac{Z^{*2} e^2}{R_0}$$

where $\frac{4\pi}{3} R_0^3 = 1/n_i$ as above.

Ion Populations

From the expressions given above, it follows that the equilibrium populations f_n are given by

$$f_n = \frac{1}{1 + \exp\left(-(\mu + I_n + \Delta I)/kT\right)}$$

where μ = electron chemical potential = $\partial F_{free}/\partial Z^*$

I_n = ionization potential = $-(1/D_n)\partial E_{ion}/\partial f_n$

ΔI = ion-sphere continuum lowering = $\partial(\Delta F)/\partial Z^*$ and Z^* is the ion charge.

These f_n can readily be shown to give the minimum total free energy, for variations at constant density and temperature.

Pressure

The pressure is now calculated form F_{tot} by differentiation with respect to the ion volume V_{ion}. In this variation, the temperature is constant; and because $\partial F_{tot}/\partial f_n = 0$ the populations f_n are also held constant. The resulting pressure contains three terms:

(1) Free electron pressure

$$p_{free} = c(kT)^{5/2}F_{3/2}(-\mu/kT)$$

where $F_{3/2}$ is the Fermi-Dirac integral

(2) A Coulomb correction

$$P_{\text{ion-sphere}} = - \partial(\Delta F)/\partial V = - \frac{9}{40\pi} \frac{Z^{*2} e^{2}}{R_{o}^{4}}$$

(3) The shell-compression pressure

$$P_{\text{shell}} = = kT \sum_{n} \log \, (1 - f_{n}) \; \partial D_{n}/\partial V$$

The Coulomb correction (2) is linked to the continuum lowering model, and the linkage is required for thermodynamic consistency. For example, if the ion-sphere expression for ΔF were replaced by the Stewart-Pyatt expression,[13] this change would induce an alteration in the Coulomb pressure expression.

The shell-compression pressure (3) is also required for thermodynamic consistency. It should be clear that this formula gives no contribution for the pressure-ionization of an unoccupied state.

Numerical calculations with the new model are shown in Figures 1-3. The figures show properties of aluminum for temperatures for 10 eV to 10 keV and densities ranging from 10^{-4} gram/cm^3 to 10^{+4} gram/cm^3. Figure 1 shows the electron pressure in Mbar; bonding corrections important near the solid state and the nuclear (ion) pressure have been omitted. Figure 2 shows the electronic energy E (Mbar-cc/gram) for aluminum under the same assumptions. Figure 3 gives the ionization state in the form of a perspective drawing.

Inspection of these results shows the new ionization model provides a smooth equation of state package. The high density behavior is considerably more realistic than that obtained from other ionization equilibrium calculations. It is planned to extend this model to provide a full range of transport and radiation coefficients.

REFERENCES

1. D. Bailey, personal communication; C. Cranfill, personal communication
2. C. A. Rouse, Prog. in High Temp. Physics and Chemistry, (Pergamon Press, Oxford, 1971), Vol. 4.
3. F. J. Rogers et al., Phys. Earth and Planetary Interiors 6, 51 (1972), Phys. Rev. A8, 1061 (1973), Phys. Rev. A10, 2441 (1974), Univ. of California preprint UCID-16776 (1975).
4. J. W. Zink, Ap. J. 162, 145 (1970); Phys. Rev. 176, 279 (1968).
5. H. C. Graboske et al., Phys. Rev. 186, 210 (1960); Phys. Rev. A3, 1419 (1971).
6. Personal communication from W. Huebner, J. Green, R. J. Olness.
7. A. McMahan and M. Ross, Lawrence Livermore Laboratory report UCRL-79015 (1977); R. M. More, Lawrence Livermore Laboratory report UCRL-50028-78-1 (1978).
8. W. Lokke, unpublished University of California preprint (1971).
9. W. Lokke and W. Grasberger, Lawrence Livermore Laboratory report UCRL-52276 (1977).
10. H. Mayer, unpublished (report LA-647 (1948)).
11. J. Green, J. Quan. Spectroscopy & Radiat. Transfer 4, 639 (1964).
12. R. M. More, unpublished calculations; A. McMahan, unpublished.
13. J. C. Stewart and K. D. Pyatt, Ap. J., 144, 1204 (1966).

FIGURE CAPTIONS

Figure 1 Electron pressure for aluminum at temperatures from 10 eV to 10 keV.

Figure 2 Electronic energy for aluminum at temperatures from 10 eV to 1 keV. The high temperature range is an ideal gas range; a shell oscillation associated with the K-shell is clearly visible at the low densities.

Figure 3 Perspective drawing of $Z^*(V,T)$ for aluminum.

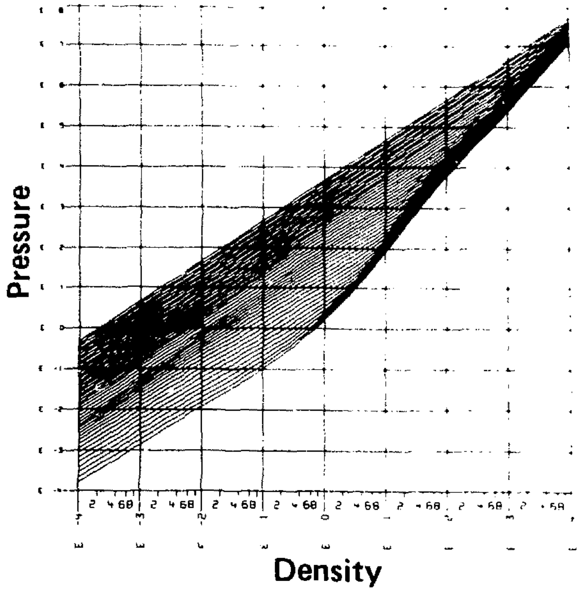

Fig. 1

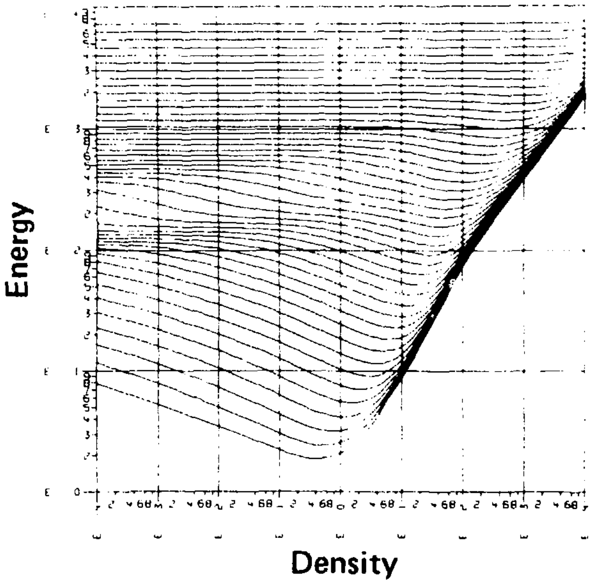

Fig. 2

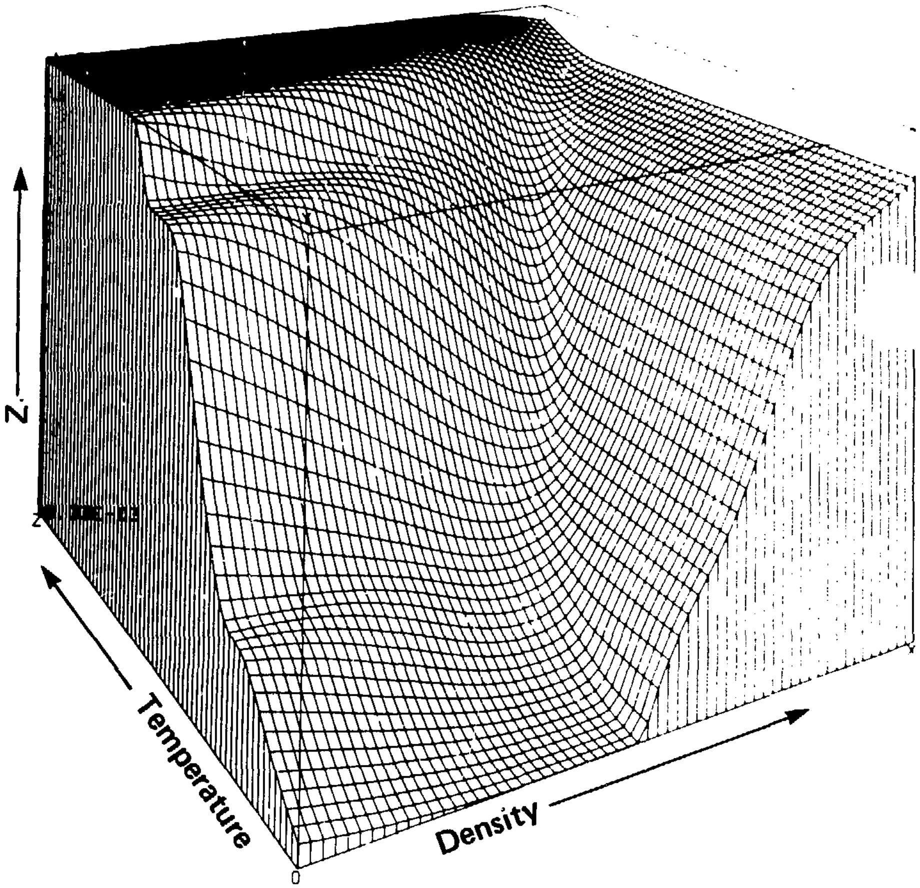

Fig. 3