

## PRESSURE IONIZATION IN LASER-FUSION TARGET SIMULATION†

G. B. ZIMMERMAN and R. M. MORE

University of California, Lawrence Livermore Laboratory, Livermore, CA 94550, U.S.A.

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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 modern laser-fusion simulation codes, this data is supplied by tables and/or calculated from a Stromgren model for ionization equilibrium. Improvements have been made in the Stromgren average-atom 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.

### 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.<sup>1-5</sup>

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.<sup>1-6</sup> For high densities, the ionization-equilibrium model typically gives results which differ significantly from the TF (Thomas-Fermi) or TFD (Thomas-Fermi-Dirac) theories. Although modest differences (10-25%) are expected from electron shell effects,<sup>7</sup> the ionization equilibrium (Saha equation) calculations disagree by as much as a factor of 2 from the TF or TFD results for high density and low temperature. In this range of temperature and density, there is good evidence that the statistical (TF, TFD) theories are more accurate.<sup>7</sup>

This report describes an improved ionization-equilibrium model which deals with these problems in a practical way. The model is based on an existing subroutine for rate equations, equation of state and opacity<sup>8,9</sup> 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.

Laser fusion simulation codes solve hydrodynamic and transport equations using physical data from a variety of sources, including analytic formulas, subroutines and tabular data. EOS (equation of state) 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%).

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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. However, the new physics in this model is introduced in an *ad hoc* fashion, with reference to the results of more elaborate quantum-mechanical calculations.

#### AVERAGE-ATOM MODEL FOR IONS

The average-atom model discussed here was introduced by Stromgren in the 1920s. It has been treated in great detail by Mayer (1948)<sup>10</sup> and Green (1965).<sup>11</sup> In recent years, it was developed as an efficient subroutine package by Lokke and collaborators at the Lawrence Livermore Laboratory.<sup>8,9</sup> We have begun from the existing subroutine and have made alterations which improve the treatment of pressure ionization.

The equations used by the code include 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 given here refer to a single element, although the code also calculates mixtures of elements.

The Stromgren model is based on a one-electron description of the atomic electron configuration; the one-electron states of an average self-consistent potential are approximately identified and each state is assigned a fractional occupation appropriate to thermal equilibrium.

The full set of hydrogenic quantum numbers  $(n, l, m, \sigma)$  or  $(n, j, j_z, \sigma)$  is truncated by assuming the energy depends only on the principle quantum number  $n$ . (This simplification only is made in the statistical portion of the code; in the opacity calculation the orbital quantum numbers are retained.) The group of electrons having fixed  $n$  is referred to as a shell; up to 10 shells are included.

The  $n$ th shell is characterized by the following quantities:  $Z_n$  = screened charge seen by electrons of the  $n$ th shell,  $\epsilon_n$  = average energy of electrons in the  $n$ th shell =  $-(Z_n^2/n^2) \times (13.6 \text{ eV})$ ,  $r_n$  = average orbit radius for electrons for the  $n$ th shell =  $(n^2/Z_n) \times (0.529 \text{ \AA})$ ,  $D_n$  = shell degeneracy =  $2n^2\phi_n(\rho)$ ; the function  $\phi_n(\rho)$  is described below,  $f_n$  = occupation = average number of electrons per quantum state in the  $n$ th 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  $n$ th shell) is  $f_n D_n$ . The screened charge  $Z_n$  is obtained from an expression of the form

$$Z_n = Z - \sum_{m \leq n} \sigma_{nm} f_m D_m,$$

where the summation includes the case  $n = m$ . The screening constants  $\sigma_{nm}$  are adopted from Ref. 10.

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

#### PRESSURE IONIZATION

The basic nature of pressure ionization can be described very simply. The electrons of the  $n$ th shell have orbit radii which are approximately given by  $r_n = (n^2/Z_n)(0.529 \text{ \AA})$ . When atoms are pushed together so that the average nuclear separation is less than  $2r_n$ , the  $n$ th shell will be severely distorted. Accurate quantum calculations show that this distortion consists of a hybridization or coupling of localized atomic states to form bands of extended wavefunctions which propagate between atoms. The width in energy of the band of extended states grows with compression and, at high compression, the bands blend into the free-electron continuum states.

Our model of these changes is simply to reduce the number of bound states of a given shell from  $2n^2$  to zero as a smooth function of density  $\rho$ . The bound electrons are thereby driven into

the continuum. The density function employed scales the reduction of  $D_n$  on the ratio  $r_n^0/R_0$ , where  $R_0 = (3/4\pi N_i)^{1/3}$  is the ion-sphere radius ( $N_i$  is the ion number density) and  $r_n^0$  is the radius of the  $n$ th shell in the neutral free atom. The scaling function is adjusted to produce reasonable results; we have examined the simple form

$$D_n = 2n^2 / \left[ 1 + \left( a \frac{r_n^0}{R_0} \right)^b \right],$$

which appears quite satisfactory.

Several comments will help to motivate this choice. First, it is intentional that  $D_n$  is calculated from  $r_n^0$  (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  $r_n^0$  are in fact not very different.

Second, the functional form chosen for  $D_n$  gradually removes the  $n$ th shell over a range of densities. This is intentional, and simulates two real phenomena in pressure ionization. One is the tendency of the low- $l$  subshells ( $s$ -states and  $p$ -states) to be pressure-ionized before the other states of a given shell.<sup>12</sup> 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.

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.<sup>2</sup> 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. The condition for thermodynamic consistency is derived from 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

$$T(\partial P / \partial T)_V = (\partial E / \partial V)_T + P$$

and this is the condition for thermodynamic consistency.

In a modern hydrodynamic code, the temperature of a fluid element is advanced according to

$$(\partial E / \partial T)_V \Delta T = -\Delta V [(\partial E / \partial V)_T + P] + \Delta U,$$

where  $\Delta U$  is the energy gained by transport and other nonhydrodynamic processes. Since  $P \Delta V$  is the change in kinetic energy, this equation shows energy conservation explicitly. For an ideal plasma,  $(\partial E / \partial V)_T + P > 0$ ; but, for very degenerate matter, the two terms are nearly equal and opposite. The use of inconsistent expressions for  $P$  and  $E$  may then cause erroneous temperature changes. The use of  $T(\partial P / \partial T)_V$  instead of  $(\partial E / \partial V)_T + P$  in the temperature equation generally yields reasonable values for  $\Delta T$ , 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 expressions are

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

$$F_{\text{ion}} = E_{\text{ion}} - TS_{\text{ion}},$$

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

$$E_{\text{ion}} = \sum_n \epsilon_n f_n D_n,$$

where the quantities  $(\epsilon_n, f_n, D_n)$  have been defined. In the expression for the ion entropy,  $k$  is the Boltzmann constant.  $F_{\text{free}}$  is the free energy of a homogeneous free electron gas (the complicated formula is available in the literature) of  $Z^*$  electrons per ion, where

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

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

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

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

## ION POPULATIONS

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

$$f_n = \frac{1}{1 + \exp [-(\mu + I_n + \Delta I)/kT]},$$

where  $\mu$  = electron chemical potential =  $\partial F_{\text{free}}/\partial Z^*$ ,  $I_n$  = ionization potential =  $-(1/D_n) \partial E_{\text{ion}}/\partial f_n$ ,  $\Delta I$  = ion-sphere continuum lowering =  $\partial(\Delta F)/\partial Z^*$ ;  $Z^*$  is the ion charge.

The  $f_n$  can readily be shown to give the minimum total free energy for variations of the populations which occur at constant density and temperature.

## PRESSURE

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

(i) The free electron pressure—

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

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

(ii) A Coulomb correction—

$$\Delta p_{\text{ion-sphere}} = -\partial(\Delta F)/\partial V = -\frac{9}{40\pi} \frac{Z^{*2} e^2}{R_0^4}.$$

(iii) The shell-compression pressure—

$$p_{\text{shell}} = -kT \sum \log(1 - f_n) \partial D_n / \partial V.$$

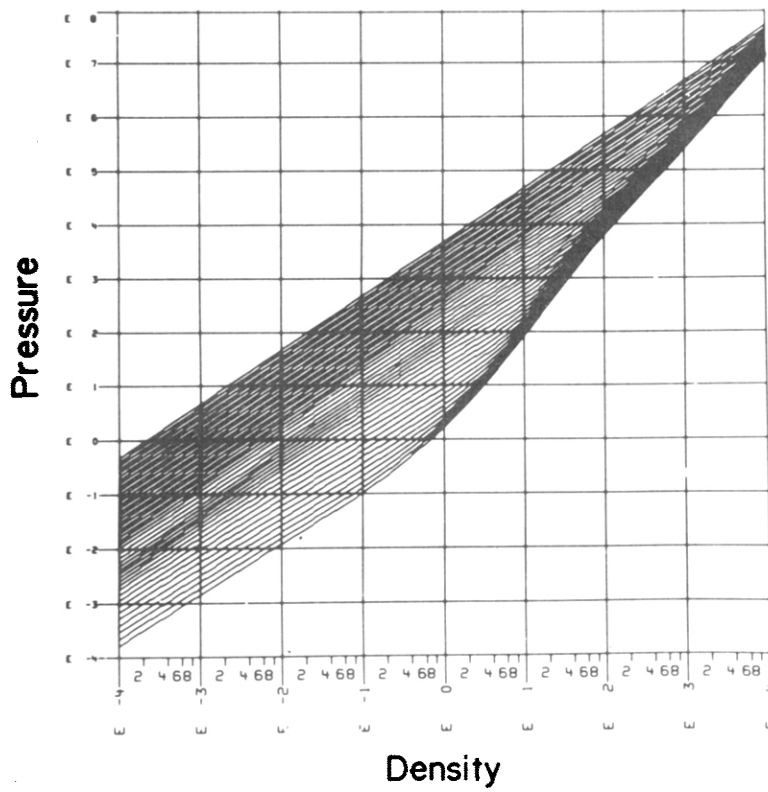


Fig. 1. Electron pressure for aluminum at temperatures from 10 eV to 10 keV.

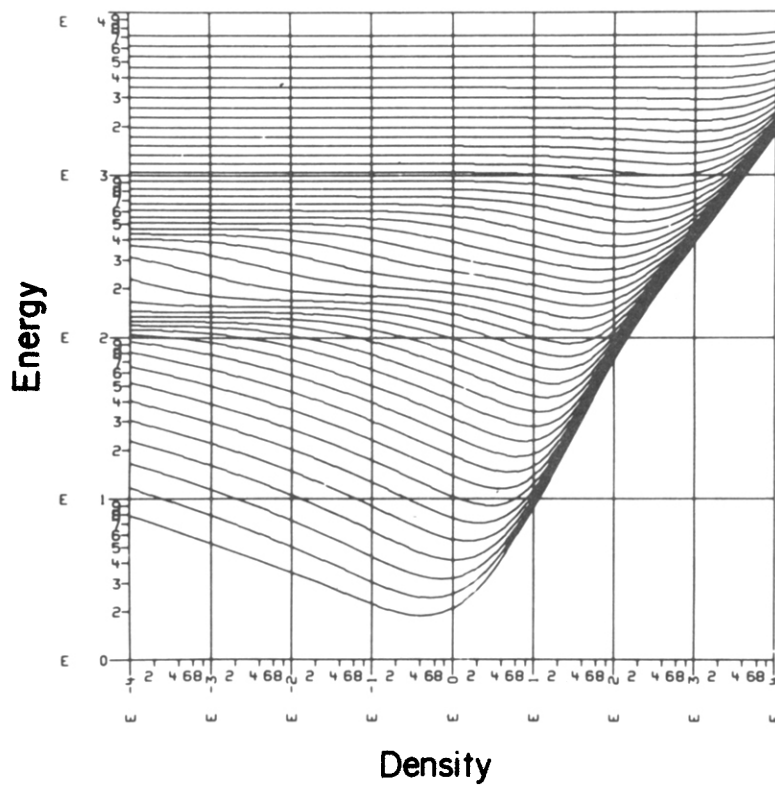


Fig. 2. Electronic energy for aluminum at temperatures from 10 eV to 1 keV. The high temperature range is an ideal gas range; a shell oscillation associated with the *K*-shell is clearly visible at the low densities.

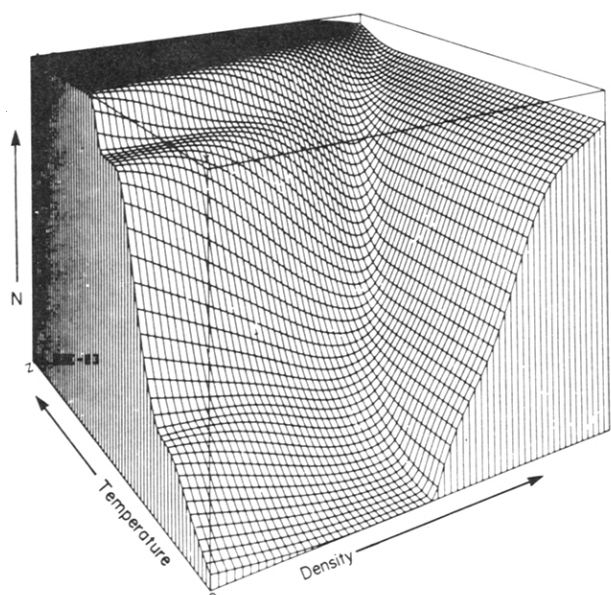


Fig. 3. Perspective drawing of  $Z^*(V, T)$  for aluminum.

The Coulomb correction (ii) is linked to the continuum lowering model, and this linkage is required for thermodynamic consistency. For example, if the ion-sphere expression for  $\Delta F$  were replaced by the Stewart-Pyatt expression,<sup>13</sup> the change would induce an alteration in the Coulomb pressure expression.

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

Numerical calculations with the new model are shown in Figs. 1–3. The figures show properties of aluminum for temperatures for 10 eV to 10 keV and densities ranging from  $10^{-4}$  g/cm<sup>3</sup> to  $10^{+4}$  g/cm<sup>3</sup>. 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. The coefficients in the equation for  $D_n$  have been selected to obtain approximate agreement with the Thomas-Fermi ionization state; the results differ by no more than 15% at any density and temperature (in fact, the 15% differences occur in the low-density range and are clearly associated with shell structure).

Inspection of these results shows that 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.

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