Approximate equation of state of some metals at high pressure from Thomas-Fermi model (*)

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Summary. — We develop on equation of state for the atom based on the elasticity formalism which extends, the validity of the Thomas-Fermi model to pressures of geophysical interest. The model is checked with shock waves data, with the Birch-Murnaghan equations and with the Earth's core equation of state.

Riassunto. — Si sviluppa un'equazione di stato per l'atomo basata sul formalismo dell'elasticità che estende la validità del modello di Thomas-Fermi a pressioni di interesse geofisico. Si fanno paragoni con i dati basati sulle onde d'urto, con le equazioni di Birch-Murnaghan e con l'equazione di stato del nucleo terrestre.

General remarks on Thomas-Fermi model.

The Thomas-Fermi [1,3] approximation was used originally for calculating the electron distribution in a heavy atom. The most important advantage of the Thomas-Fermi model is that it requires the solution of a total differential equation, subject to initial and boundary conditions, rather than partial differential equation subject to boundary conditions implying eigenvalues of constants of the motion for many electrons, as in Schrodinger equation. Thus it constitutes essentially an approximate method of solution of the many-electron

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problem, analogous to the Debye-Huckel theory ([4,5]) of ionic solutions. The method has been applied to the approximate calculation of many atomic properties. The basic idea of the Thomas-Fermi approximation is to consider the electrons classically provided that they satisfy the exclusion principle. Then the electron density can be calculated by means of the Poisson equation. The atom is divided into cells small enough so that the potential is essentially constant, but large enough so that the electrons in a cell occupy a region of phase space large compared with h^3. At 0°, the Fermi-Dirac statistics implies that the electron density in the phase space is $2/h^3$ at all points corresponding to an energy less than the Fermi energy, and is zero at points corresponding to an energy larger than the Fermi energy. At a given point of the configuration space, those points of the momentum space within a sphere of radius corresponding to the maximum momentum permitted by the Fermi energy, are occupied by electrons, and those outside the sphere are unoccupied. The total number of electrons within the sphere determines the electron density which is to substitute in Poisson's equation and, by solving this equation, one finds the potential and the electron distribution self-consistently. Besides the statistical approach, it is also possible to derive the Thomas-Fermi model as the semiclassical limit of the Hartree equations for the model of a self-consistent central field ([6,7]). Since the sublevels of angular momentum are averaged, it is possible to describe only those properties of the atom which are independent or little dependent of the shell structure. At high pressure such structure must be largely destroyed, so that the equation of state at the highest compressions should be accurate.

From an opposite point of view, we must recall also that the Thomas-Fermi model has some defects and we review the most interesting trial corrections available up today.

The electron density becomes infinite as $1/r^{3/2}$ at the nucleus and vanishes as $1/r^6$ at large distances; this is in contradiction with the behaviour of the wave mechanical electron density which is constant at the nucleus and vanishes exponentially at large distance from the nucleus. Also the binding energies, as calculated from Thomas-Fermi theory, are 10-30% lower than the empirical values.

Many authors have corrected the original model using more exact forms for the potential and the kinetic energy. To eliminate the self-interaction of the electrons from the electrostatic coulomb interaction, the only one take into account in the original model, Fermi and

Amaldi [8] have subtracted from the potential of the atom the mean potential V_e/Z, of one electron, where V_e is the potential of the complete electron cloud and Z the atomic number. This correction gives a good approximation at the border of the atom and improves the electron distributions in this region.

Moreover Dirac [9] took into account the exchange interaction between electrons determining a fundamental change of the boundary electron distribution: the radii of the atoms become finite, i.e. the electron density vanishes on the border.

Another correction comes from considering [10,11,12] the correlation energy resulting from the interaction correlation of the electrons. This is smaller than the other two corrections mentioned above and plays an important role only in the border of the atom.

Various corrections for the kinetic energy are also considered. First is the so-called Weizsacker [14] correction, by which the singularity of the electron density at the nucleus and the $1/r^6$ falling off of it at infinity disappear. However the agreement with the empirical data is not so good: the energies of the model are 20-25% higher than the experimental values. Moreover this model cannot be deduced in a completely satisfactory manner [15,16,17].

The most satisfactory correction is due to Plaskett [18] obtained starting from the Schrodinger equation.

However, concluding those general considerations, it appears, as pointed out by Gombas [10], that one of the most important problems in the statistical theory of atoms is the completely satisfactory deduction of the kinetic energy correction. A further task would be the deduction from wave mechanics of a general relation between electron density and potential which would include the kinetic energy correction in a natural way, and would hold even for the lightest atoms. Strictly speaking, this would not simply mean a further development of the statistical theory itself, but, rather, the deduction of this general wave-mechanical relation in an adequate approximation. The point is just the "adequate approximation". The exact relation is extremely involved; the point would really be to deduce a useful approximation from the general exact relation, which would satisfy the physical conditions.

The present work is concerned exclusively with the application of Thomas-Fermi model to the calculation of an equation of state at high pressure. For other applications, to atoms and to nuclei, the works of Gombas [13,19] can be consulted.

II. – The domain of applicability.

An equation of state inferred from the statistical atom model becomes more accurate the larger the atomic number Z and the higher is the pressure P. The first condition follows from the requirement that the number of electrons be large enough in a cell in the atom, for which the potential is relatively constant over an electron wavelenght, so that the corresponding region occupied in phase space be large relative to h^3. The second condition is a consequence, on the basis of the assumptions of the model, since it corresponds to the condition under which the detailed structure of the energy levels can be ignored. However, we must remember that at low pressure, the effects of chemical binding and lattice structure of the solid are ignored in the statistical atom model. These effects account for binding in the solid state at normal pressure, and the procedure of viewing the effect of the lattice merely as a perturbation of properties inferred from the statistical model is valid only at high pressure.

It was emphasized by Feynman, Metropolis and Teller ([1]) that the equation of state inferred from the statistical atom model are likely to be valid only for pressures exceeding about 10 Mb. Elsasser ([20]) has suggested a few million megabars, beginning at somewhat higher pressures for the lighter elements and somewhat lower for the heavier elements.

This question of the critical pressure can now be examined on the basis of measurements of shock waves in metals. The original measurements by Walsh and Christian ([21]) for aluminium, copper, and zinc for shock pressures up to about 500 kb were extended by McQueen and Marsh ([21]) to many elements up to a pressure of 2 Mb. In subsequent work by Altschuler et al ([23]), the upper limit of pressure has reached 5 Mb. The data for pressure versus density of Altschuler and al. and of McQueen and Marsh have independently been reduced to zero absolute temperature by Takeuchi and Kanamori ([24]) and compared with the predictions of the Thomas-Fermi Dirac theory. They find that the Thomas-Fermi model yields densities that are too low at 10 Mb, but extrapolation of the experimental results indicates that the actual equation of state is well represented by the model at pressures slightly higher than 100 Mb. The conclusion applies to Ag, Au, Cd, Cu, Fe, Pb, and Zn, involving extrapolations in pressure by a factor 20 to 50, in general. On the basis of the results of Takeuchi

and Kanamori, a critical pressure of about 100 Mb, independent of atomic number, seems the most reasonable.

Until the advent of the data from measurements on shock waves, laboratory determinations of equations of state could be carried out to pressure of only 0.1 Mb as an upper limit, by the techniques of Bridgman ([25]). The procedure of interpolating the equation of state at intermediate pressures from the computation of the statistical model at high pressure and the measurements of Bridgman at low pressure has been used for geophysical purposes by Elsasser ([20]), Bullen ([26]), Birch ([27]) to infer the chemical composition of the core. Nevertheless data from shock-wave measurements now extend to 5 Mb, in excess of the pressure (3-6 Mb) at the Earth's Center, this method, has its merits as pointed out by Gilvarry ([28]), in spite of obvious deficiences. The Elsasser's conclusion that the core is composed of iron with a possible mixture of nickel and the deduction by Knopoff and MacDonald ([29]) that the iron of the core is probably alloyed with silicon, obtained with use of this interpolation procedure, have not been reversed by the direct experimental data obtained from the shock-wave measurements.

Only with use of such an interpolation scheme the equation of state from the statistical atom model can be applied with any validity to the planets of the solar system, since the maximum pressure in the interior (for Jupiter) is only about 30 Mb, less than the critical pressure 100 Mb adopted above. However, pressures considerably higher than this critical value occur in the interiors of white dwarf stars. Thus the equation of state from the statistical atom model can be applied in the determination of the mass-radius relation and the limiting maximum mass for white dwarf stars (Chandrasekhar ([30])).

As an example of the application of results from the statistical atom model to a question in planetary science, the problem of predicting the pressures and temperatures arising in explosive impact of large meteorites on the surface of the Moon and Earth can be discussed for astronomic meteorite velocities. This was treated by Gilvarry and Hill ([31]).

It is desiderable to indicate schematically the domain of applicability of the statistical atom model in the field of the variables, temperature T (absolute) and pressure P. This is done in Figure 1, adapted from a corresponding diagram of Wares ([38]) in terms of temperature and density by Gilvarry ([28]) and with correction to the atomic number of iron in the non-relativistic case and with addition of regions corres-

ponding to condensed phases. The plane of the diagram is separated into two disjoint regions by the locus corresponding to the degeneracy temperature; for temperatures below this curve, Fermi-Dirac statistics must be used to characterize the electrons while, for temperatures above, the Fermi-Dirac statistics reduce to the usual Maxwell-Boltzmann form.

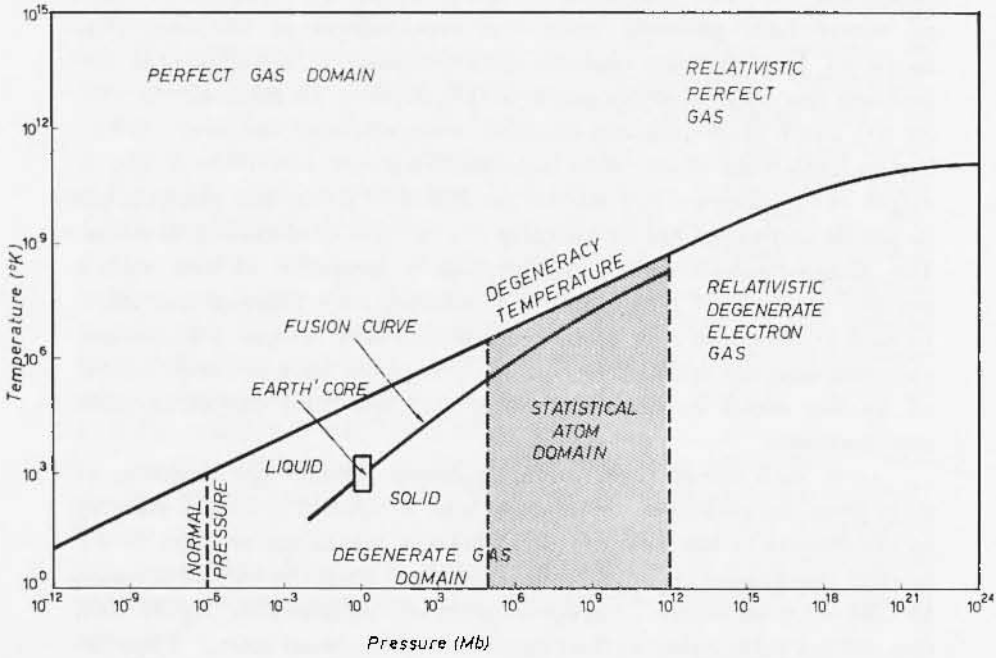

Figure 1. – Schematic representation of the domain of applicability of the statistical atom model in the fields of variables, temperature T (absolute) and pressure P. The statistical atom model is approximately valid at temperatures below the degeneracy temperature, at pressures below that where relativistic effects become important, and at pressures above a limit of about 100 Mb necessary for the assumptions of the model to be valid; the corresponding region of the variables is shown cross-hatched. The lines for the degeneracy temperature and the fusion curve (at higher pressures) as shown correspond roughly to iron, and in general should be represented by bands for a range of atomic number. The rectangle indicates the coordinate point for the Earth's. Adapted from Wares (for the relativistic case) with modifications and additions for the non-relativistic region by Gilvarry.

Erratum: The line that represents the fusion curve does not cross the statistical atom domain.

The statistical atom model to be described in what follows applies to temperatures below the degeneracy locus, at pressures above the critical

pressure of about 100 Mb discussed above but below the pressure at which relativistic effects become important. The degeneracy temperature appearing is approximately that for iron, and the corresponding domain of validity of the statistical atom model is shown cross-hatched. This region is cut into two parts by the fusion curve, separating the regions of existence of the liquid and solid phases. The melting line as drawn for the higher pressures corresponds roughly to iron. For coordinate points below this curve, corrections to thermodynamic functions as inferred from the statistical atom model are necessary for the effect of the lattice. One notes that the fusion curve does not fall far from the degeneracy locus; thus the domain of validity of the non-relativistic statistical model to be described is roughly coextensive with the region of the solid phase. Actually the degeneracy transition from Fermi-Dirac to Maxwell-Boltzmann statistics is continuous but not sharp and this, as well as the other boundaries of region in Figure 1, corresponds to ranges of atomic number. Thus, all lines of demarcation in Figure 1 should be drawn as bands, hence the corresponding numbers appearing on the coordinate axes are indicative of orders of magnitude only. It should be noted that the degeneracy criterion specified applies only the electrons, while the heavy atomic particles (protons, neutrons and nuclei) follow the classical Maxwell-Boltzmann statistics over essentially the entire field of the diagram.

The coordinate point corresponding to the Earth's core is shown in Figure 1 by a rectangle whose height represents roughly the uncertainty in the temperature as inferred by Gilvarry [33,34,35]. Normal pressure is indicated by a vertical broken line. It should be noted that the free electrons in a metal are strongly degenerate under conditions of normal pressure and temperature.

The purpose of this work is to modify the Thomas-Fermi equation of state to extend its validity to the range of pressures of geophysical interest. This will be made by considering the atom, in such a range of pressures, formally as an elastic body.

III. – The equation of state.

The central question is the determination of the equation of state of materials at extreme conditions is the calculation of the electronic configuration of the system. In the Thomas-Fermi theory each atom of the material occupies an independent spherical cell, and the electron

distribution is determined to a first approximation about a nucleus fixed in the center of the cell. As already mentioned, the electrons are assumed to be free Fermi-Dirac particles, all other details of the quantum mechanics of atoms are ignored and the distribution of the electron cloud is related to the electrostatic potential by Poisson's equation. In this manner the main effects of Coulomb interactions are included self-consistently to all orders in the electric charge. Then, by classical kinetic theory, neglecting the (small) contribution of the nuclear motion to the thermodynamics of the system, it is possible to derive the equation of state.

In the case of neutral spherically atoms, there is no electric field at the atomic boundary. All of the momentum carried across the surface of the atom is due to the kinetic energy electrons. On the basis of the kinetic theory of a free electron gas, we can write:

$$p(r_o) = \frac{2}{3} c_o \varrho^{5/3} (r_o) \qquad [1]$$

with

$$c_o = \frac{3 h^2}{10 m} \left(\frac{3}{8 \pi} \right)^{2/3}$$

where ϱ is the electron density, p the pressure and r_o the atomic radius. Calculating ϱ from the Thomas-Fermi equation, it is possible to obtain the equation of state.

Now let us assume that [1] is valid for all the values of

$$r_o : 0 \leqslant r_o \leqslant r_o (p = 0)$$

i.e. let:

$$p(r) = \frac{2}{3} c_o \varrho^{5/3} (r) \qquad [2]$$

give the behaviour of the pressure in the interior of the atom. Then let us write the following relation formally derived from classical theory of elasticity:

$$\tau = \Delta \lambda \qquad [3]$$

where τ has the meaning of an external perturbation (applied pressure), λ is the reaction (internal pressure) to the external perturbation and Δ

is the dilatation. Then assuming $p(r) = \lambda(r)$ the surface displacement is given by

$$s = \left\{ \frac{1}{r_o^2} \int_o^{r_o} \frac{r^2}{p(r)}\, dr \right\} \tau \qquad [4]$$

we can derive the following equation of state:

$$\frac{\varrho_1 - \varrho_o}{\varrho_1} = \left\{ \frac{3}{r_o^3} \int_o^{r_o} \frac{r^2\, dr}{p(r)} \right\} \tau_1 \qquad [5]$$

where ϱ_o is the initial density and ϱ_1 is the density after first compression τ_1. If the atom undergoes other subsequent compressions we can write generally

$$\frac{\varrho_n - \varrho_{n-1}}{\varrho_{n-1}} = \left\{ \frac{3}{r^3{}_{n-1}} \int_o^{r_{n-1}} \frac{r^2\, dr}{p(r)} \right\} (\tau_n - \tau_{n-1}) \,. \qquad [6]$$

In the equations [5] and [6], $p(r)$, given by [2], has the following explicit form:

$$p(r) = A\,(Z) \left| \frac{\Phi\,(x)}{x} \right|^{5/2} ; \qquad A\,(Z) = \frac{e^2 Z^2}{10\, \mu^4\, \pi} \qquad [7]$$

obtained from Thomas-Fermi model. In the equation [7] Z is the atomic number, Φ is the Thomas-Fermi function, e is the electron charge, x is a dimensionless variable:

$$x = \frac{r}{\mu} ; \qquad \mu = 0.88 \cdot a_o \quad Z^{-1/3} \text{ cm}$$

where a_o is the Bohr radius for hydrogen.

Substituting [7] in [6] and putting:

$$I_{n-1} = \int_o^{x_{n-1}} \frac{x^{9/2}}{\Phi^{5/2}(x)}\, dx \qquad [8]$$

we obtain

$$\frac{\varrho_n - \varrho_{n-1}}{\varrho_{n-1}} = \left\{ \frac{3}{x^3{}_{n-1}\ A(Z)}\, I_{n-1} \right\} \tau_n \,. \qquad [9]$$

Using the input parameters given by Table 1, we have developed numerical calculation for Fe, Cu, Zn, Ag, Cd, Au, Pb considering two subsequent compressions (the former, from 0 to 10^{12} dynes/cm^2, the latter from 10^{12} to $4 \cdot 10^{12}$ dynes/cm^2) and the results are shown by dashed curves, in Figg. 2-8.

Table 1. – Input parameters for numerical calculations (Royce [37]).

Metal	Z	$\frac{\varrho_0}{(\mathrm{g/cm^3})}$	$\frac{r_0}{(\mathrm{\AA})}$
Fe	26	7.86	1.61
Cu	29	8.90	1.41
Zn	30	7.14	1.54
Ag	47	10.49	1.60
Cd	48	8.64	1.73
Au	79	19.24	1.59
Pb	82	11.34	1.93

Comparisons with shock waves data.

Altshuler et al. [22] have measured the compressibility of several metals to a pressure of the order of $4 \cdot 10^{13}$ dynes/cm^2 using the technique of shock waves. The equation of state so determined must be reduced to a reference temperature (0° K) in order to make proper interpretations of the experimental results. This has been made in two different manners by Knopoff and MacDonald [26], and by Takeuchi and Kanamori [24]. Figg. 2-8 show the results of Takeuchi and Kanamori for Fe, Cu, Zn, Ag, Cd, Au, Pb compared with Thomas-Fermi Dirac equation of state, with [9] and with the Birch-Murnaghan [27] equation of state for three values of the parameter ξ. The results of Knopoff and MacDonald for Fe, Cu, are represented in Fig. 9 and compared with [9], the Thomas-Fermi equations of state, and the p-V relation of Earth's interior. From these considerations we can conclude that our model constitutes an improvement of the equations of state derived from the Thomas-Fermi model of the atom and that the formal use

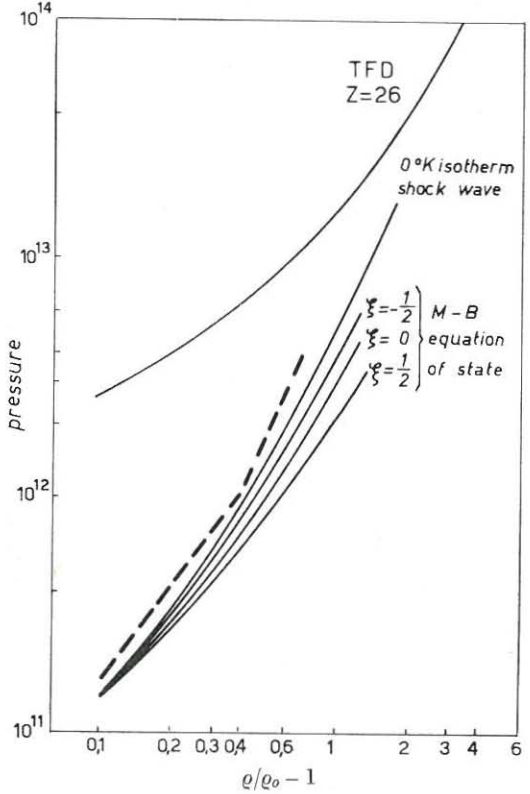

Figure 2. – Isotherms of iron at 0° K based on shock wave data reduced by Takeuchi and Kanamori [21], Birch-Murnaghan model and Thomas-Fermi model. The dashed curve represents our model.

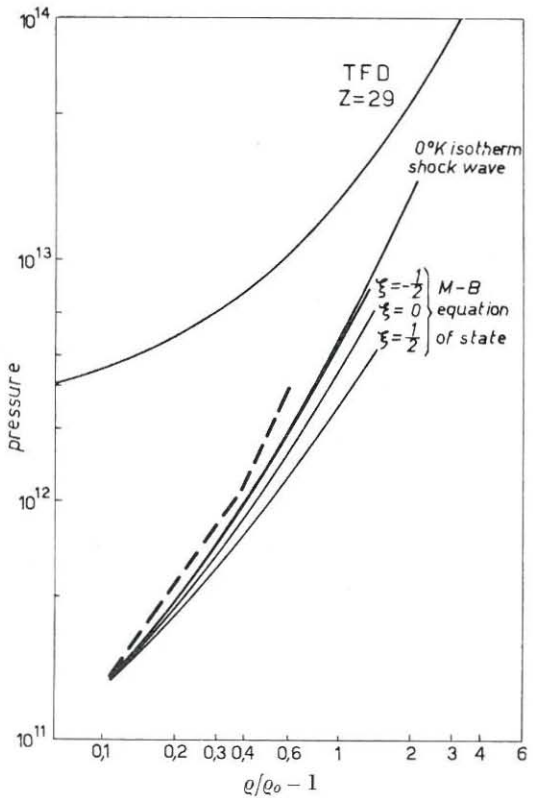

Figure 3. – Isotherms of Copper, at 0° K based on shock wave data reduced by Takeuchi and Kanamori [24], Birch-Murnaghan model and Thomas-Fermi model. The dashed curve represents our model.

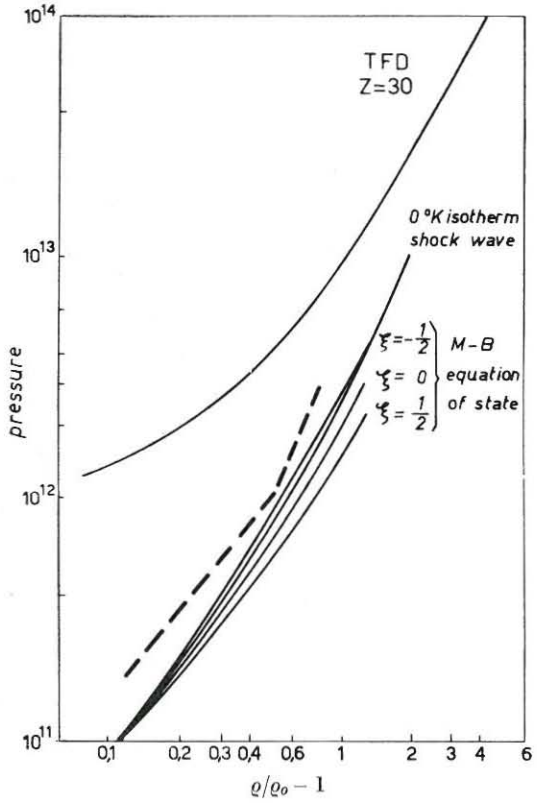

Figure 4. – Isotherme of Zinc at 0° K based on shock wave data reduced by Takeuchi and Kanamori (24), Birch-Murnaghan model and Thomas-Fermi model. The dashed curve represents our model.

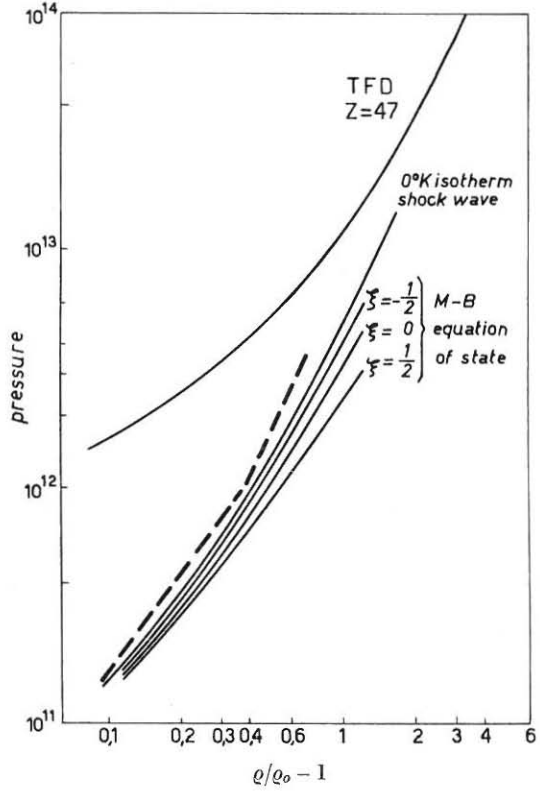

Figure 5. – Isotherme of Silver at 0° K based on shock wave data reduced by Takeuchi and Kanamori (24), Birch-Murnaghan model and Thomas-Fermi model. The dashed curve represents our model.

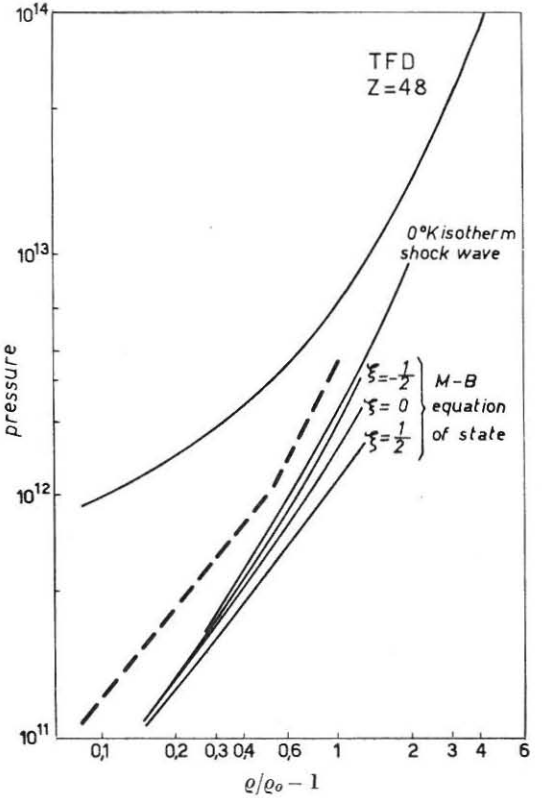

Figure 6. – Isotherme of Cadmium at 0° K based on shock wave data reduced by Takeuchi and Kanamori [24], Birch-Murnaghan model and Thomas-Fermi model. The dashed curve represents our model.

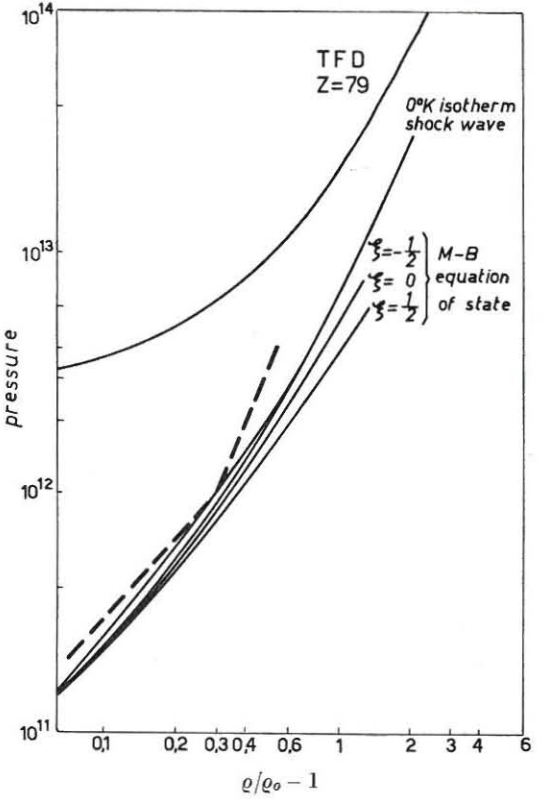

Figure 7. – Isotherme of Gold at 0° K based on shock wave data reduced by Takeuchi and Kanamori [24], Birch-Murnaghan model and Thomas-Fermi model. The dashed curve represents our model.

of elasticity in atomic scale is justified from the experimental results. However we want to note that the present results are only estimates and that in a next work we shall report a more rigorous development

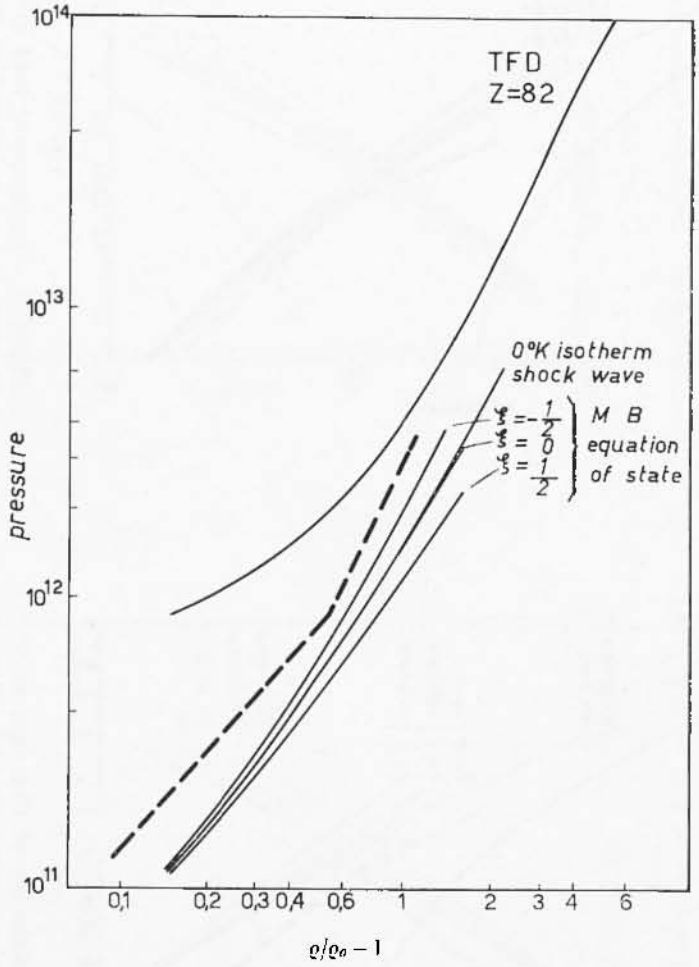

Figure 8. – Isotherme of Lead at 0 °K based on shock wave data reduced by Takenchi and Kanamori [24], Birch-Murnaghan model and Thomas-Fermi model. The dashed curve represents our model.

and more precise results. The temperature perturbation also shall be taken into account.

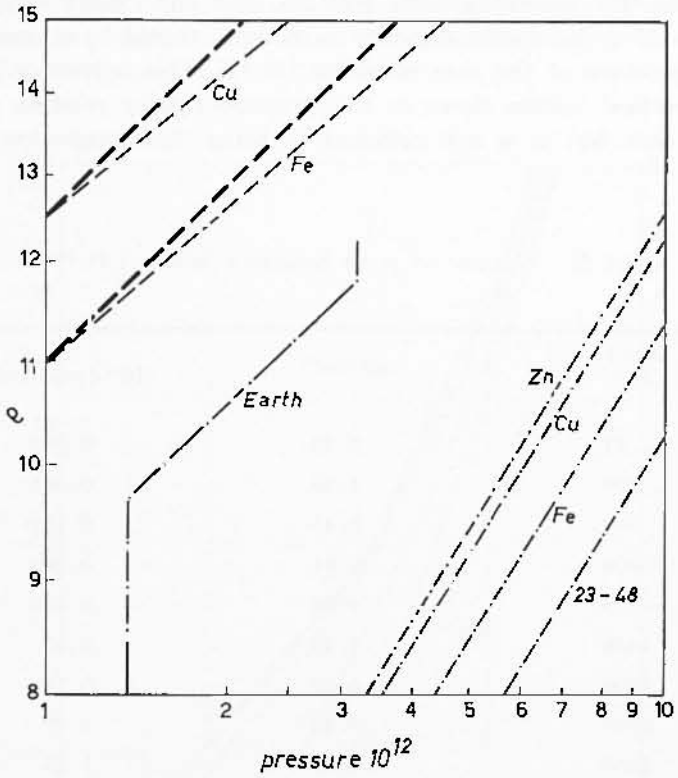

Figure 9. – Equations of state for iron, copper, zinc, and a hypothetical material of atomic number 23, atomic weight 48 in the Earth's core pressure range. The values derived from shock wave measurements (solid) are compared with those obtained from the Thomas-Fermi theory (full circles). Bullen 's density distribution is shown for comparison (triangles). Our results are indicated by dashed lines.

Geophysical Implications

Our results for iron are compared with the Earth's core equation of state. For this purpose we use the pressure-density relation of the Earth's interior given by Bullen's model (A[1]) ([36]) reported in Table 2.

We must note first that it is very significant that the theoretical curve of iron almost coincides with the laboratory experiments (Fig. 2). It is also significant that our theoretical curve of iron is much closer

to the Earth's pressure-density relation than the former theoretical models. Also the thermodinamic corrections should be at most 8% for temperature of the core of about 10^4 °K. This correction brings the theoretical results closer to the pressure-density relation of the Earth's core, but it is not sufficient to bring them toghether.

Table 2. – VALUES OF p, IN BULLEN'S MODEL (A¹) ([38]).

Depth (Km)	(gs/cm³)	p (10^{12} dynes/cm²)
33	3.32	0.009
200	3.36	0.064
400	3.41	0.132
600	4.01	0.206
1000	4.66	0.383
1400	4.90	0.57
1800	5.12	0.77
2200	5.33	0.98
2600	5.54	1.20
2700	5.59	1.26
2883	5.68	1.37
2883	9.79	1.37
3000	9.97	1.49
3500	10.65	1.99
4000	11.19	2.45
4500	11.60	2.84
4982	11.89	3.15
6371	12.22	3.55

Therefore the equation of state introduced in this paper, when compared with the pressure-density relation for the Earth's interior, can suggest anomalies which are of physical significance. For instance,

if we accept the hypothesis of a core made of iron and silicates whose representative atomic number is 23, then the relative composition should be 80% iron and 20% silicates.

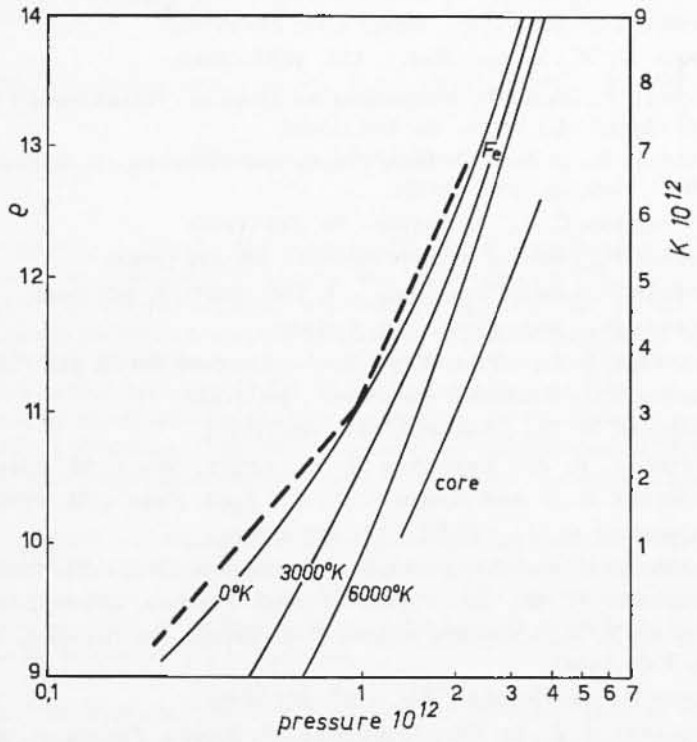

Figure 10. – Comparison of density curves for iron with those for the Earth's core. Density distribution is that of Bullen's model A. Our results are represented by dashed lines.

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