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Generalized Thomas–Fermi approach for systems under pressure

E. Cappelluti^{a,*}, L. Delle Site^b

^a*Dipartimento di Fisica, Università “La Sapienza”, P.le Aldo Moro 2, 00185 Roma, Italy*

^b*Max-Planck Institute for Polymer Research, Ackermannweg 10, D 55021 Mainz, Germany*

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Abstract

In a previous work one of the authors proposed a simple model for studying systems under pressure based on the Thomas–Fermi (TF) model of single atom. In this work we intend to extend the previous work to more general TF models where electronic exchange and correlation are introduced. To do so, we first study numerically the equation obtained by Lewis (TFDL) which introduces the effects of exchange and correlation into the original TF equation; next the procedure followed in the previous work is extended to the new approach and a specific example is illustrated. Although one could expect that no big differences were produced by the generalized TF model, we show the qualitative as well as quantitative equivalence with detailed numerical results. These results support the robustness of our conclusions with regards to the model proposed in the previous work and give the character of universality (i.e., to pass from one atom to another, the quantities calculated must be simply scaled by a numerical factor) to the properties of compressed systems shown in this work. © 2002 Elsevier Science B.V. All rights reserved.

1. Introduction

The statistical model of atom developed by Fermi [1] and known as Thomas–Fermi (TF) model, although based on a highly simplified theoretical framework, has been proven surprisingly good in predicting basic properties of condensed matter [2]. A particular feature of such a model is the description of compressed atoms, in which case the predicted properties were confirmed experimentally [3]. In a preceding publication [4], we focused the attention on this particular aspect of the theory and proposed a simple model for describing systems under pressure. The central point of that work was the development of the concept of “statistical ionization”. In simple terms, this

* Corresponding author.

E-mail address: emmcapp@pil.phys.uniroma1.it (E. Cappelluti).

is a universal semianalytical function which enables one to describe, as a function of the distance from the point-like nucleus, the balancing process between the antibinding and binding contribution to the total energy within the compressed atom. In spite of the approximations done and extensively discussed, we underlined the utility of the proposed model as a tool for investigating at a basic level and at low computational cost, properties of systems under pressure. However it was also underlined that the properties of semianalyticity and universality of the function disappear when higher order of approximation are introduced in the basic TF theory. In the light of what stated before, in this publication we intend to extend the treatment of the previous work to more sophisticated models of the TF approach. That is to say, to include the effects of exchange and correlation into the original TF model and obtain in this framework a “generalized statistical ionization function”. The extension of the original TF model is due to Dirac (TFD) [5] for the exchange while for the part relative to the correlation, several approaches has been proposed; in this work the one proposed by Lewis (TFDL) [6] has been chosen because it is simple and appropriate for the compressed case, since the treatment of the electrons becomes exact in high density limit (see the appendix). The paper is organized as follows; first we obtain the TF, TFD and TFDL equation within a single generalized approach, then we show numerical solutions for different atomic numbers in the neutral uncompressed case, next we illustrate numerical results for the “generalized statistical ionization function” in case of $Z = 50$ (Z atomic number). Finally the equation of state of a compressed system is calculated. Comments on the results obtained as well as on advantages and limitations of this model, conclude the work.

2. A statistical approach: the Thomas–Fermi–Dirac–Lewis (TFDL) model

In this section we derive a unique general form for the TF, TFD and TFDL equation. In a semiclassical approximation the local electronic density of states can be defined as (see for example Ref. [7]):

$$dn[p(\mathbf{r}), (\mathbf{r})] = \frac{8\pi}{h^3} p^2(\mathbf{r}) dp(\mathbf{r}). \quad (2.1)$$

The local electron density is therefore given by

$$n(\mathbf{r}) = \int_0^{p_F(\mathbf{r})} \frac{8\pi}{h^3} p^2(\mathbf{r}) dp(\mathbf{r}) = \frac{8\pi}{3} \frac{p_F^3(\mathbf{r})}{h^3}, \quad (2.2)$$

which determines the local Fermi vector $p_F(\mathbf{r})$.

In the spirit of the (generalized) Thomas–Fermi approach we can express the one-particle energy $E[p(\mathbf{r}), \mathbf{r}]$ through an effective Hamiltonian $H_{el}[p(\mathbf{r})]$ of free (interacting) electrons plus an electrostatic field $V(\mathbf{r})$ arising from the direct electron–nucleus and electron–electron interaction (we must remind that the nucleus is considered a positive point-like charge):

$$E[p(\mathbf{r}), \mathbf{r}] = H_{el}[p(\mathbf{r})] - eV(\mathbf{r}). \quad (2.3)$$

For a system at the equilibrium the chemical potential $\mu(\mathbf{r})$, defined by the maximum energy of $\max\{E[p(\mathbf{r}),(\mathbf{r})]\} = E[p_F(\mathbf{r}),\mathbf{r}]$, has to be a constant μ independent of \mathbf{r} . Putting for convenience $\mu = -eV_0$ we obtain therefore

$$H_{\text{el}}[p_F(\mathbf{r})] = e[V(\mathbf{r}) - V_0]. \quad (2.4)$$

The effective field $V(\mathbf{r})$ will be determined self-consistently via electrostatic considerations by using the Poisson's equation

$$\nabla^2[V(\mathbf{r}) - V_0] = \frac{1}{r} \frac{d^2}{dr^2} \{r[V(r) - V_0]\} = e \frac{n(r)}{\varepsilon_0}, \quad (2.5)$$

where $r = |\mathbf{r}|$. Eq. (2.5) can be quite simplified by introducing the variables $\phi(r)$ and $v(r)$ defined by

$$\frac{Ze}{4\pi\varepsilon_0 r} \phi(r) = V(r) - V_0, \quad (2.6)$$

$$v^2(r) = (2Ze^2 m)^{-1} (4\pi\varepsilon_0) r p_F^2(r). \quad (2.7)$$

Eq. (2.5) becomes thus

$$\phi''(r) = \frac{1}{Z\varepsilon_0} \left(\frac{8\pi}{3h^3} \right) (2ZE^2 m)^{3/2} (4\pi\varepsilon_0)^{-1/2} v^3(r) r^{-1/2}, \quad (2.8)$$

and finally, by scaling $r \rightarrow \beta a_0 x$, we have

$$\phi''(x) = v^3(x) x^{-1/2}. \quad (2.9)$$

This is the “generalized TF equation”, where

$$a_0 = \frac{4\pi\varepsilon_0 h^2}{4\pi^2 m e^2} \quad (2.10)$$

is the atomic Bohr radius and

$$\beta = \left(\frac{9\pi^2}{128Z} \right)^{1/3}. \quad (2.11)$$

A closed system of differential equations is now obtained by expressing back the variable $\phi(x)$ as function of $v(x)$. To this aim the explicit expression of the electronic Hamiltonian $H_{\text{el}}[p(r)]$ is needed. Of course this task is in principle quite hard since the solution of a many-body system is necessary, and some kind of approximation is required. The original TF model represents the simplest approximation, i.e., the Hamiltonian $H[p(r)]$ is approximated with the only kinetic term

$$H_{\text{el}}^{\text{TF}}[p(r)] = \frac{p^2(r)}{2m}. \quad (2.12)$$

The total Hamiltonian contains therefore only kinetic and the direct Coulomb electron–electron interaction, totally neglecting any quantum contribution. By using again the reduced variables $\phi(x)$ and $v(x)$, we can write Eq. (2.12) in the compact form

$$\phi(x) = v^2(x), \quad (2.13)$$

which, together with Eq. (2.9), defines the statistical solution of the Thomas–Fermi model.

As stated before higher order of approximation have been introduced in literature to include quantum exchange and correlation contribution to Eq. (2.12). Most famous is the so called TFD model which explicitly takes into account the exchange energy (see Refs. [5,2]). The inclusion of the correlation term has been a more debated issue, and different approaches have been proposed. A particular suitable and simple one is the interpolation formula given by Lewis, which becomes exact in the both the high-density and low-density limits [6]. We referred and will refer to it as the TFDL model; in this case the Hamiltonian contains kinetic, exchange and correlation terms

$$H_{\text{el}}^{\text{TFDL}}[p(r)] = \frac{p^2(r)}{2m} - \frac{e^2 p(r)}{4\pi\epsilon_0\pi\hbar} - \frac{me^4(1 - \ln 2)}{(4\pi\epsilon_0\pi\hbar)^2} \ln \left[1 + \frac{(0.89\alpha\pi - 1)\pi p(r)a_0}{(1 - \ln 2)\hbar} \right], \quad (2.14)$$

where

$$\alpha = \left(\frac{4}{9\pi} \right)^{1/3}. \quad (2.15)$$

Introducing the variables $\phi(x)$ and $v(x)$, Eq. (2.14) reads

$$\phi(x) = v^2(x) - av(x)x^{1/2} - \rho a^2 x \ln[\sigma a + v(x)x^{-1/2}], \quad (2.16)$$

where

$$a = \left(\frac{2\beta}{\pi^2 Z} \right)^{1/2}, \quad (2.17)$$

$$\sigma = \frac{1 - \ln 2}{2(0.89\alpha\pi - 1)}, \quad (2.18)$$

and

$$\rho = \frac{1}{2}(1 - \ln 2). \quad (2.19)$$

Eq. (2.16) can be considered a generalization of the TF model [Eq. (2.13)] to include exchange and correlation energies. We find Lewis's formula particularly appealing since we are mainly interested in the high-density regime of compressed atoms where the interpolation formula provided by Eq. (2.14) is expected to work quite well. In spite of the simplicity of the model, to our knowledge, no explicit numerical or analytical treatment of this equation has been published, although several authors [8–11] cite this approach in their work. The main reason why this happens is due to the fact that since the TFD model does not produce evident changes with respect to the simple TF solutions, the TFDL is expected not to be much different.

However in our paper we derive for the first time explicit numerical solutions of the TFDL model and we use it as basic starting point of our analysis; the results will

be compared with the simple TF model. Although we do not expect large differences with respect to the TF, we carry on this study to confirm a qualitative and quantitative equivalence between TF and TFDL and later on the consequent advantages of such a conclusion will be underlined.

3. Numerical study of TFDL equations and “generalized ionization function”

In this section we illustrate some TF, TFD and TFDL solutions for the neutral uncompressed case, the trend shown for this case is preserved for the compressed case. Then we show within a generalized TF approach, the procedure to obtain the “statistical ionization function” via the total energy as a function of the distance from the nucleus. Eqs. (2.9) and (2.16), or Eqs. (2.9) and (2.13), represent two sets of coupled differential equations which can be solved to obtain $\phi(x)$ or, equivalently, $v(x)$, and as a consequence the local electron density $n(r)$. The boundary conditions required to determine a unique solution are provided by the asymptotic limit:

$$\lim_{r \rightarrow 0} \phi(x) = 1, \quad (3.1)$$

and by fixing the radius of a neutral atom

$$\phi'(x_0) = x_0 \phi(x_0). \quad (3.2)$$

Although it has been widely pointed out in literature, it is important to remind that the inclusion of exchange and correlation terms breaks down the universality of the solution. Namely, it is not possible to obtain local electron density $n(r)$ for a given atomic number Z just by a proper scaling of the physical quantities (length, energy, etc.). In Fig. 1 we show the radial distribution of $\phi(x)$ for a neutral uncompressed atom for the Thomas–Fermi model and for the Thomas–Fermi–Dirac–Lewis model with statistically relevant values of atomic number: $Z = 10, 50, 80$. Only a slight discrepancy is found as a result of the inclusion of the exchange and correlation terms, pointing out the almost quantitative accuracy of the TF model. As expected, such a discrepancy vanishes as the atomic number Z is increased. This result shows that TF and TFDL are quantitatively as well as qualitatively equivalent (the trend as stated before holds in the compressed case as well).

By the knowledge of the solution $\phi(x)$ we can now evaluate the semiclassical expression of the local density of energy $E(r)$ which is related to the one electron energy $E[p(r), r]$ through the relation $E(r) = \int dn[p(r), (r)] E[p(r), r]$. We can identify three different contributions:

$$E(r) = E_{\text{kin}}(r) + E_{e-i}(r) + E_{e-e}(r), \quad (3.3)$$

where

$$E_{\text{kin}}(r) = \int dn[p(r), (r)] \frac{p^2(r)}{2m}, \quad (3.4)$$

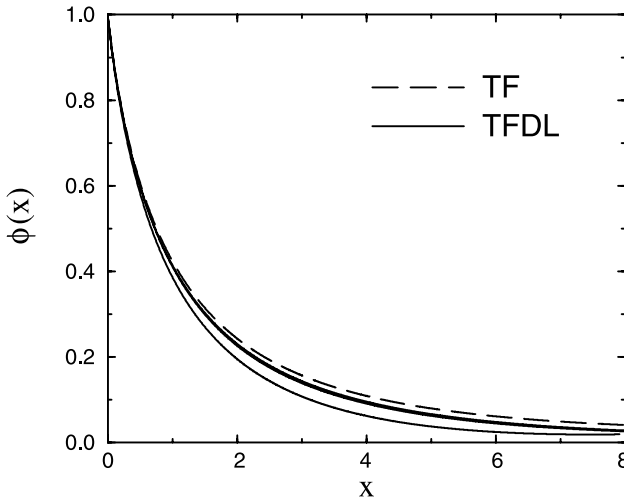


Fig. 1. Radial distribution of $\phi(x)$ as function of x for a neutral uncompressed atom. Dashed line represents the Thomas–Fermi model, solid lines the Thomas–Fermi–Dirac–Lewis one with atomic number (from lower to upper line) $Z = 10, 50, 80$. The $Z = 50$ and 80 cases are strongly overlapping and are hardly distinguishable in the plot.

$$E_{e-n}(r) = - \frac{Ze^2}{4\pi\epsilon_0 r} n(r), \tag{3.5}$$

$$E_{e-e}(r) = \frac{1}{2} \frac{e^2}{4\pi\epsilon_0} n(r) \int_{|\mathbf{r}'| < r_0} \frac{d\mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|}, \tag{3.6}$$

where of course

$$n(r) = \int dn[p(r), (r)]. \tag{3.7}$$

A particularly meaningful quantity is also the integrated energy defined $E(R)$ by

$$E(R) = \int_0^R 4\pi r^2 dr E(r), \tag{3.8}$$

where R is a generic distance $R \leq r_0$. $E(R)$ represents therefore the total energy contained in the atomic volume enclosed within the distance R from the nucleus.

Just as $E(r)$, we can think $E(R)$ as composed by a kinetic, a Coulomb electron–nucleus and a Coulomb electron–electron term. After some straightforward manipulations (see also Ref. [12]), by using once more the reduced variables, we can write each of these contributions as

$$E_{\text{kin}}(X) = \frac{2^{10/3}(3\pi)^{1/3}}{5\pi} Z^{7/3} \left[\frac{me^4}{2(4\pi\epsilon_0\hbar)^2} \right] \int_0^X dx v^5(x) x^{-1/2}, \tag{3.9}$$

$$E_{e-n}(X) = - \frac{2^{10/3}(3\pi)^{1/3}}{3\pi} Z^{7/3} \left[\frac{me^4}{2(4\pi\epsilon_0\hbar)^2} \right] \int_0^X dx v^3(x) x^{-1/2}, \tag{3.10}$$

$$E_{e-e}(X) = \frac{1}{2} \frac{2^{10/3}(3\pi)^{1/3}}{3\pi} Z^{7/3} \left[\frac{me^4}{2(4\pi\epsilon_0\hbar)^2} \right] \int_0^X dx v^3(x) x^{1/2} \times \left[\frac{1}{x} \int_0^x v^3(x') x'^{1/2} + \int_0^{x_0} v^3(x') x'^{-1/2} \right], \tag{3.11}$$

where X is the scaled variable related to R through the relation $R = \beta a_0 X$ and where we made use of the relation:

$$\int_{|\mathbf{r}| < R} \int_{|\mathbf{r}'| < r_0} \frac{f(\mathbf{r})f(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}' = \int_0^R dr 4\pi r^2 f(r) \left[\frac{1}{r} \int_0^r dr' 4\pi r'^2 f(r') + \int_0^{r_0} dr' 4\pi r' f(r') \right], \tag{3.12}$$

valid for an isotropic $f(\mathbf{r})$.

As was done in Ref. [4], we consider R a variable distance within the atom, and $E(R)$, as stated before, is the total energy in classical terms, of the sphere of radius R inside the spherical atoms. In simple terms, we do divide the atom into infinitesimally thin concentric shells; the energy at R is the sum of the contributions of all of the shells inside R . As a consequence the distance at which $E(R)$ has its minimum can be interpreted as the distance at which the binding (nucleus–electrons) and antibinding (electron–electron and kinetic energy) contributions to the total energy are in exact balance. This allows us to define a sort of electron ionization criterion, where the term “ionization” stands for electrons with zero or positive energy; thus we can address to $E(X)$ as the “generalized statistical ionization function”. The exact meaning of “statistical ionization” with the related limitations has been extensively discussed in our previous work, for such a reason we do not spend more discussion about it and focus the attention onto the numerical results.

The number of ionized electron is therefore given by

$$N_{\text{ion}} = \int_{R \leq |\mathbf{r}| \leq r_0} n(\mathbf{r}) d\mathbf{r}, \tag{3.13}$$

which, written in reduced variables, reads

$$N_{\text{ion}} = Z \int_{x_{\text{core}}}^{x_0} dx v^3(x) x^{1/2}. \tag{3.14}$$

Differently from the procedure adopted in our previous work, in this case $E(X)$ can be studied only numerically, since it is not possible to solve analytically the integrals and so obtain a semianalytical expression. Moreover the dependence of $\phi(x)$ on Z does not allow to obtain a universal function where to pass from one atom to another is possible by simply scaling in Z . Next, we show results obtained by studying numerically the “generalized statistical ionization function” for the case $Z = 50$; however since we did find quantitative agreement between TF and TFDL, the results shown are valid for any atomic number by opportunely scaling them.

4. Results

In this section we show results obtained for the “generalized statistical ionization function” for the case $Z = 50$. As it was already underlined, we expect that the results are quantitatively and qualitatively equivalent in case TF or TFDL model is used, since the solution of the TF and TFDL equation does not differ, as it was shown in the previous section. This means that the “generalized ionization function” is the numerical equivalent of the semianalytical and universal “statistical ionization function” of our previous work; thus the results (and the equation of state) shown here for $Z = 50$, can be easily generalized to any Z by simply scaling. Indeed the function $E(X)$ does not show a different behavior when TF or TFDL are used; in Fig. 2 the plots obtained using TF and using TFDL coincide. As it is possible to see in Fig. 2 a minimum is always obtained for any compression, and in Fig. 3 a particular compression has been chosen and the determination on the number of ionized electrons is pictorially illustrated.

Accordingly to the procedure of our previous work, at this point we can therefore model the compressed atom as a core with radius R_{core} plus a number of ionized electrons N_{ion} which, in principle, is free and is spread over all the atomic volume. The pressure is therefore determined by the density of “free” (ionized) electrons as we proposed in the previous work (again limitations are extensively discussed there):

$$P = \frac{(3\pi^2)^{2/3}}{5} \frac{\hbar^2}{m} \left(\frac{N_{\text{ion}}}{V_{\text{atom}}} \right), \tag{4.1}$$

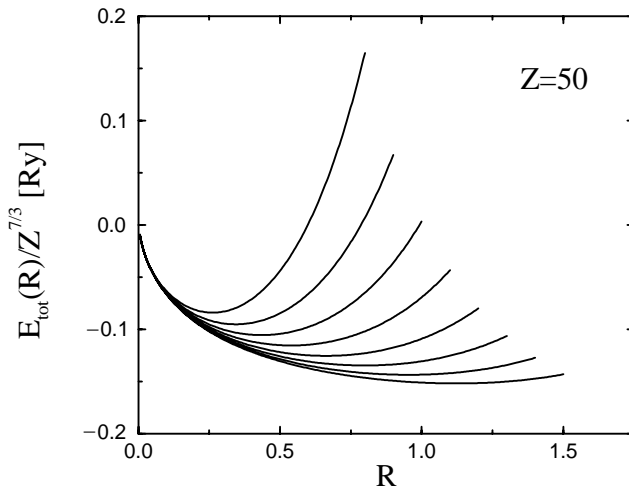


Fig. 2. Integrated energy $E(R)$ as function of R for the Thomas–Fermi–Dirac–Lewis model ($Z = 50$) and different degrees of compression parameterized by the atomic radius (from lower to upper line): $x_0 = 1.5, 1.4, 1.3, \dots, 0.9, 0.8$. $E(R)$ always presents a minimum at a certain R .

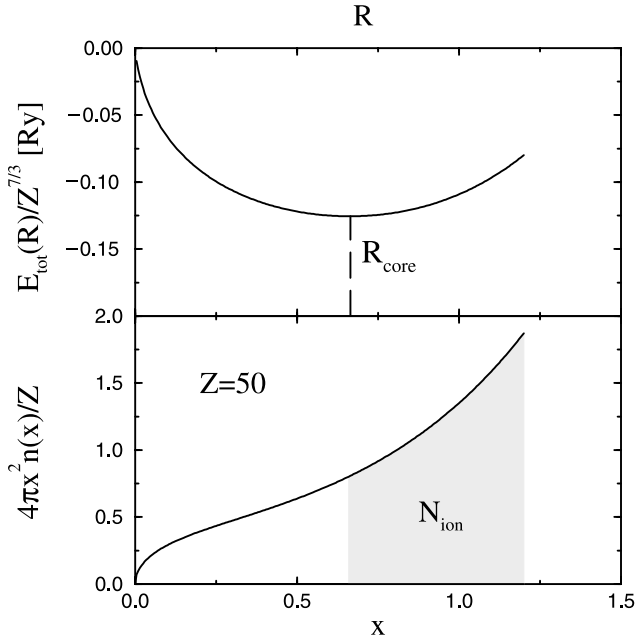


Fig. 3. Top panel: particular of Fig. 2 for $x_0 = 1.2$. The minimum of $E(R)$ determines the “core” radius R_{core} . Bottom panel: graphical representation of the number of ionized electrons N_{ion} corresponding to the above case. N_{ion} is determined by integrating the electron density from the “core” radius to the atomic boundary.

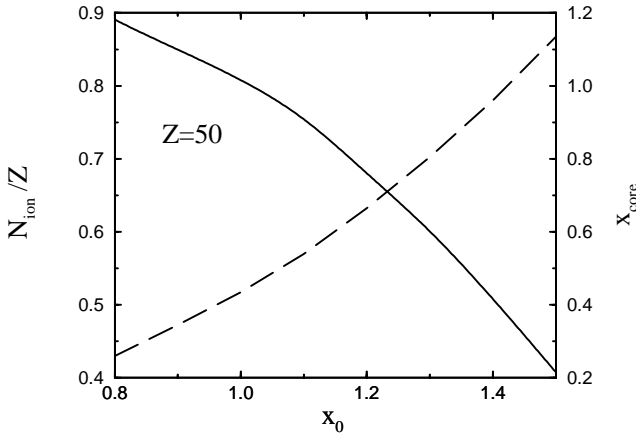


Fig. 4. Number of ionized electrons (solid line, left scale) and core radius x_{core} (dashed line, right scale) as function of the atomic radius x_0 .

where N_{ion} is given by Eq. (3.14) and the atomic volume is simply $V_{\text{atom}} = r_0^3$. First we study the ionization as a function of the compression for the single atom (see Fig. 4) and then we extend the procedure to a macroscopic level so that the resulting equation

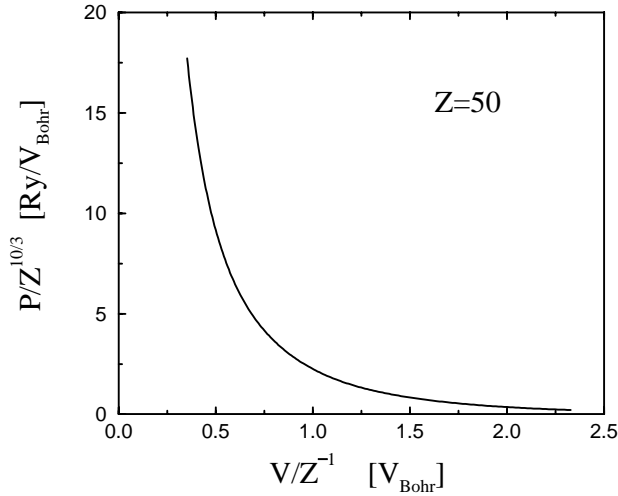


Fig. 5. Pressure vs. volume dependence for the statistical ionization model.

of state is shown in Fig. 5, where the pressure in units of Rydberg over the Bohr atomic volume $V_{Bohr} = a_0^3$ is plotted as function of the volume.

5. Discussion and conclusion

We generalized the concept of “statistical ionization function” for electrons in a compressed atom, obtained in a previous work, within the TF approach. The generalization was developed by extending the previous analysis to more sophisticated TF models where exchange and correlation are considered. We found that there is no qualitative and quantitative difference between the original and the generalized approach. Although one could have expected this result, we explicitly proved such an equivalence, and for the first time, solutions of the TFDL equation were shown. The important conclusion of this work concerns the fact that because of the equivalence shown, the results reported for the particular case of $Z = 50$ are valid for any atomic number, provided that a rescaling $Z^*/(Z = 50)$, where Z^* is the atomic number wished, is applied; above all the results legitimate the use of the semianalytical and universal ionization function obtained in our previous work with the evident advantages of the low computational cost and its extreme simplicity and immediacy. Moreover the fact that Lewis’ formula is exact for the high density limit, means that in our case the effects of correlation were well described, as a consequence one can conclude that such effects are not relevant for describing atoms under pressure, at least in first approximation; this result it is not obvious in an a priori analysis. As it was discussed in the previous work, the equation of state is a simplification and as a consequence far from being rigorous; for instance an open problem of our model is the distribution of the “ionized” electrons which in

the present work is considered simply uniform. However, due to its simplicity and feasibility, the model can be applied for a basic study of compressed matter not only via the determination of the equation of state, but also as a basis for developing more efficient analysis within more sophisticated theories. The example shown in our previous publication, was the determination of the “ionized” electrons for a certain compression and the consequent description, in an *ab initio* method, of the ionized electrons via plane-waves wavefunctions while the other electrons can be represented as a core or as localized orbitals; this would speed up the convergence for self-consistent calculations of compressed matter. Here we can say more, as it is well known, the *ab initio* calculations are based on the concept of pseudopotential; only the valence electrons are explicitly taken into account while the rest are placed in a core described by an opportune potential which interacts with the valence electrons. Valence electrons and core are known for the uncompressed atoms but one may ask what happens in case the system is under high pressure, in this case the valence electrons and the core should be redefined according to the degree of compression. In this case our model ‘at basic level’ would be very helpful for estimating the number of ionized (valence) electrons and define what is the core. The evident advantage of such a procedure stays in the simplicity of such an estimate and in the very low computational cost. In conclusion we think that the study performed in this work furnishes important information and tools for a computational inexpensive and well founded basic analysis of compressed matter.

Appendix A. Correlation energy within Lewis’ approach

We briefly illustrate the procedure followed by Lewis to develop a suitable formula for the electron correlation, for more details see Ref. [6] and references therein. Consider an electronic Fermi gas in an effective field $V(\mathbf{r})$, with an electron density modulated through $p(\mathbf{r})$. The correlation energy at high density can be calculated via the Gell–Mann’s scheme and leads to the following expression:

$$H^{\text{corr}}[p(\mathbf{r})] = - \frac{me^4(1 - \ln 2)}{(4\pi\epsilon_0\pi\hbar)^2} \ln[p(\mathbf{r})] + \text{const} , \quad (6.1)$$

where m and e are, respectively, the electron mass and the electron charge. This expression must be modified in such a way that its validity could be reasonably extended to any density and in particular must be exact at low density. At this point Lewis invokes Wigner procedure for the calculation of electron correlation for a dilute gas; i.e., one simply needs to note that a very dilute electron gas in the ground state crystallize into a body-centered cubic lattice and at this point the correlation energy can be determined exactly via a Madelung type technique. The expression for low density obtained is

$$H^{\text{corr}}[p(\mathbf{r})] = - \frac{(0.89\alpha\pi - 1)e^2 p(\mathbf{r})}{4\pi\epsilon_0\pi\hbar} , \quad (6.2)$$

where $\alpha = (4/9\pi)^{1/3}$. Finally the procedure leads to what Lewis defines as “a suitable interpolation formula” for the correlation energy valid for any density:

$$H^{\text{corr}}[p(\mathbf{r})] \approx -\frac{me^4(1 - \ln 2)}{(4\pi\epsilon_0\pi\hbar)^2} \ln \left[1 + \frac{(0.89\alpha\pi - 1)\pi p(\mathbf{r})a_0}{(1 - \ln 2)\hbar} \right], \quad (6.3)$$

where $a_0 = 4\pi\epsilon_0\hbar^2/me^2$ is the Bohr radius. This is the expression used by Lewis to incorporate the correlation effects into the TFD model.

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