Extended Thomas-Fermi Models for Equations of State of Dense, High Energy-Density Matter

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Our goal is to develop an equation of state model for dense, high energy-density matter—which has densities near solid and pressures exceeding one megabar—that spans temperatures from tens of electron volts to several kilovolts. Density functional theory (DFT) is our starting point, since it is formally exact and therefore allows us to construct rigorous, self-consistent models of wide applicability. In the high-density and/or high-temperature limit, we know that DFT tends toward the so-called Thomas-Fermi (TF) model; unfortunately, solid density is too low for this limit to be of use. Moreover, there is evidence that the first obvious correction to basic TF theory, which adds an exchange term to the free energy and carries the name "Thomas-Fermi-Dirac" (TFD), can actually be worse [1, 2]. It is also generally recognized [2] that the most important correction to TF theory is the inclusion of the cusp condition near a nucleus, which makes the electronic density finite there. The cusp arises from a gradient correction term known as the "Weizsäcker" (TFDW) correction, which occurs with various coefficients **λ**. To understand these issues we are working to develop a *rigorous* formulation of extended Thomas-Fermi theory that is an all-electron theory, does not employ ad hoc scaling factors **λ** of the gradient terms, and does not assume that various physical effects (e.g., kinetic energy and exchange) enter in an additive manner; that is, our formulation will be internally self-consistent.

To understand the main features that we wish to keep in our formulation, we have first studied various forms of extended TF theory. Specifically, we have studied cold, neutral atoms because (1) we can study variations with nuclear charge, (2) it is possible to compare with very accurate experimental and theoretical results, and (3) the low temperature regime represents a difficult limit of our intended high-temperature regime of interest. As we are also interested in a computationally efficient

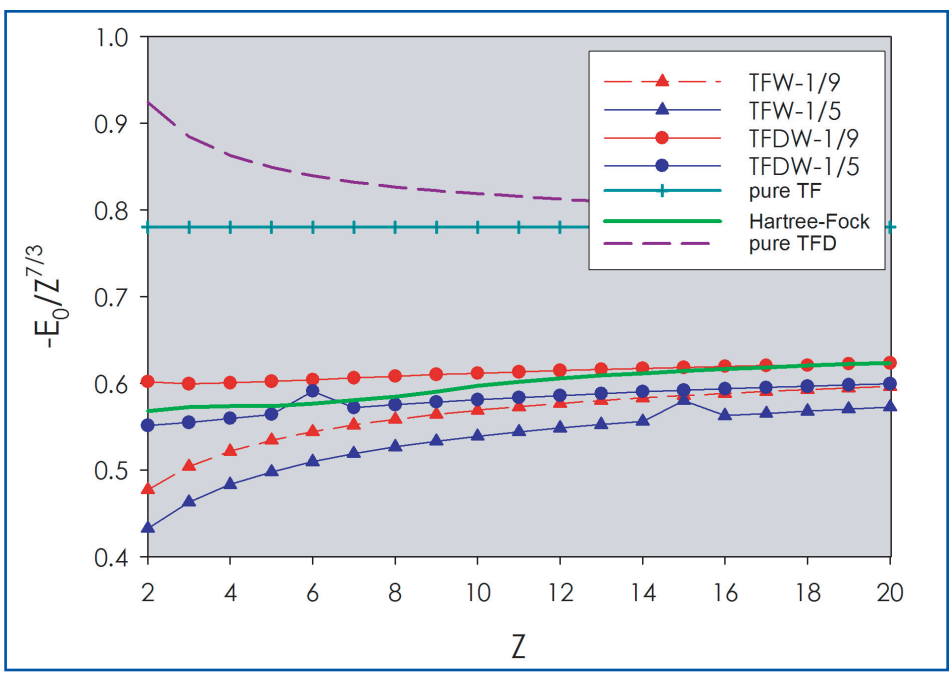
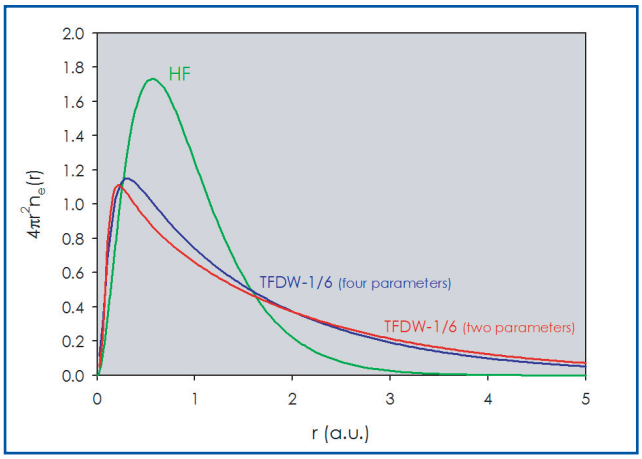


Figure 1— ***This figure shows the ground state energy of atoms, in terms of $-E_0/Z^{7/3}$, versus nuclear charge Z as computed with several models. The most important extension is the gradient correction (TFDW). The fractions following the TFDW results are the values of the coefficient λ. The Dirac (exchange) (TFD) correction to TF degrades the result.***

method, we also use the simple case of atomic systems to develop a variational method to find the electronic density. In addition to computational efficiency, our variational method employs an electronic density that has desirable behaviors, such as a finite value near the nucleus. We choose various forms for the parameterization to ensure convergence. In practice, we construct forms for the free energy that correspond to different extended TF theories, all of which depend only on the density, and perform a minimization with respect to the parameters of the density subject to normalization of the density. We have used two different methods for performing the minimization, and we report results in this paper that use the Powell method.

We display some of our results in Figs. 1–3. In the first figure we show the ground state energy of atoms with nuclear charges Z = 2-20 as computed from various models, including the accurate Hartree-Fock (HF) theory. We plot $-E_0/Z^{7/3}$, where E_0 is the actual ground state energy, because this quantity is constant in pure TF theory. Our results confirm that TF is not very accurate, TFD is worse, and models with gradient correction that are consistent with the cusp condition are quite good with about 10% agreement with HF. Fig. 2 shows the actual density for the case of helium (Z = 2) and poor agreement is found, as expected for a low-Z element. This indicates that other quantities might be more or less sensitive to the result depending on how they weight the density variations. To quantify the density variations we compare density moments in Fig. 3 between HF and our parameterized density versus nuclear charge. For higher-Z elements, the moments are quite well described with extended TF models.

In conclusion, we feel that an extended TF model will yield accurate equations of state for hot, dense matter provided exchange ("Dirac") and gradient terms are included rigorously and self-consistently. Although other contributions, such as correlation corrections beyond the mean-field, may be important at high temperatures [3], we feel that we will be able to obtain extremely accurate equations of state. For dense matter we will develop our "high temperature-DFT" first in a spherical cell and then in a macrocell consisting of many—possibly different—nuclei. We have made considerable progress in developing such a formulation from DFT, and this will be reported in the future.



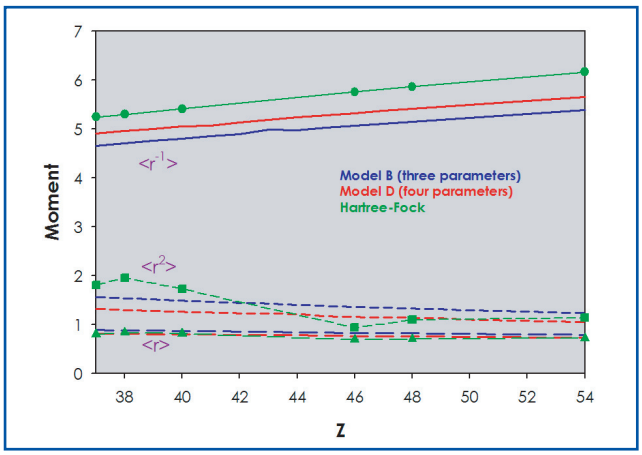


Figure 2— ***Here we show the radial density $4\pi r^2 n(r)$ for helium from three models: green (HF), red (our TFDW-1/6 with two parameter minimization), and blue (same as red with four parameters). The agreement is poor, as expected for a low-Z element.***

Figure 3— ***Moments of the density are shown versus nuclear charge from several models: green (HF), blue (three parameter TFDW), and red (three parameter TFDW). Overall agreement between extended TF and HF is good and convergence with respect to the number of parameters is seen.***

[1] J.F. Barnes and R.D. Cowan, *Phys. Rev.* **132**, 236 (1963).
[2] R.G. Parr and W. Yang, *Density-Functional Theory of Atoms and Molecules* (Oxford University Press, Oxford, 1994).
[3] U. Gupta and A.K. Rajagopal, *Phys. Reports* **87**, 259 (1982).