International Journal of Thermophysics, Vol. 13, No. 2, 1992

Shell Effects in the Equation of State of Metals

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Received June 20, 1991

A simple quasiclassical Z-scaling model (QMT) is proposed to calculate the electron equation of state (EOS) for matter with a high energy concentration. This model may be employed over a wide range of densities and temperatures from the Saha model region of application to the corrected Thomas–Fermi model model (TFC) area of use. The treated model describes ab initio typical step behavior of the ionization state and energy as a result of successive shell ionization with increased temperature. The model naturally includes the effects of electron–ion interaction with increased density. This EOS model may be interesting for gas dynamics, energetics, astrophysics, etc.

KEY WORDS: equation of state; high pressures; ionization; quasiclassical approach; Thomas–Fermi model.

1. INTRODUCTION

In this paper the quasiclassical electron equation of state (EOS) is proposed for matter with a high energy concentration. The statistical EOS models [1] commonly employed in this range of parameters are the Thomas–Fermi (TF) model [2], the TF model with gradient and exchange corrections (TFK or TFC) [3, 4], and the quantum-statistical model [5–7], treating these corresctions self-consistently.

The statistical TF model is a dominant term of the Hartree–Fock model expansion in a semiclassical parameter $\xi = d\lambda\!\!\!^{-}/dx$ ($\lambda\!\!\!^{-}$ is de Broglie wavelength). It averages all the physical quantities ignoring the atomic shell structure. The gradient and exchange corrections have a second order in ξ and do not change the averaged behaviour of physical functions. However, a Taylor series in ξ cannot describe all the atomic system

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features because the functions are nonanalytical in ξ. There are terms $\sim\xi^n \exp(i/\xi)$ and a separation of the dominant such term enables us to consider the shell effects [8] and to construct the EOS quasiclassical model.

It should be noted that some semiclassical aspects (wave functions, quantization condition) one can find in different publications, for example, in Refs. 9–11. But if one uses a succesive semiclassical approach, all the resulting physical quantities are the terms of an expansion series in a semiclassical parameter ξ, the dominant TF term, the gradient, exchange, and shell corrections being included. A similar approach has been used independently in Ref. 12 to describe the total binding energy of an isolated atom at zero temperature.

2. GENERAL PRINCIPLES

The TF model in a Wigner–Seitz spherical cell is the basis of our theory. Electron density, particle number, free energy, internal energy, and pressure in the TF model are (in the following equations the atomic units are used)

$$\rho_{\mathrm{TF}}(r)=\frac{\sqrt{2}}{\pi^2}\,T^{3/2}I_{1/2}\left(\frac{\mu-V(r)}{T}\right) \tag{1}$$

$$N_{\mathrm{TF}}=\int d\bar{r}\,\rho_{\mathrm{TF}}(r) \tag{2}$$

$$F_{\mathrm{TF}}=-\frac{2\sqrt{2}}{3\pi^2}\,T^{5/2}\int d\bar{r}\,I_{3/2}\left(\frac{\mu-V(r)}{T}\right) -\frac{1}{2}\int d\bar{r}\,\rho_{\mathrm{TF}}(r)\left[V(r)+\frac{Z}{r}\right]+\mu N \tag{3}$$

$$E_{\mathrm{TF}}=\frac{\sqrt{2}}{\pi^2}\,T^{5/2}\int d\bar{r}\,I_{3/2}\left(\frac{\mu-V(r)}{T}\right)+\frac{1}{2}\int d\bar{r}\,\rho_{\mathrm{TF}}(r)\left[V(r)-\frac{2Z}{r}\right] \tag{4}$$

$$P_{\mathrm{TF}}=\frac{2\sqrt{2}}{\pi^2}\,T^{5/2}I_{3/2}(\mu/T) \tag{5}$$

where T, μ, Z, and $I_n(x)$, respectively, are the temperature, chemical potential, nuclear charge, and Fermi-Dirac integral, and $V(r)$ is a potential electron energy,

$$V(r)=-\frac{Z}{r}+\int d\bar{r}'\,\frac{\rho_{\mathrm{TF}}(r')}{|\bar{r}-\bar{r}'|}$$

If one wants to account for some other effect in addition to the TF approach, there is a general expression for the correction to the TF free energy through some effect [1]:

$$\delta F = -\int_{-\infty}^{\mu} d\mu' \, \delta N(\mu') \tag{6}$$

where $\delta N(\mu') = \int d\bar{r} \, \delta\rho(\bar{r}, \mu')$ is the particle number correction, the correction to the TF electron density $\delta\rho$ through the effect being small.

In the treated nonrelativistic self-consistent potential approach, the TF-method does not consider the effects of the electron density inhomogeneity, exchange interaction, and atomic shell structure. The use of Eq. (6) leads us to construct the additive expression for the free energy with regard to all the above effects on the TF-model basis. The result is a quasiclassical EOS model (QM):

$$F_{\mathrm{QM}} = F_{\mathrm{TF}} + \delta F_{\text{qu-ex}} + \delta F_{\mathrm{sh}} \tag{7}$$

$$E_{\mathrm{QM}} = E_{\mathrm{TF}} + \delta E_{\text{qu-ex}} + \delta E_{\mathrm{sh}} \tag{8}$$

$$P_{\mathrm{QM}} = P_{\mathrm{TF}} + \delta P_{\text{qu-ex}} + \delta P_{\mathrm{sh}} \tag{9}$$

where subscripts "qu-ex" and "sh" denote quantum-exchange and shell. Since quantum-exchange corrections are calculated as in the TFK model [4], in this paper we consider the shell corrections only.

One can derive from Eq. (6) the pressure correction in a general form,

$$\delta P = -\left.\frac{\partial \delta F}{\partial W}\right|_{T} = \rho_{\mathrm{TF}}(R_0)\, \delta\mu + \int_{-\infty}^{\mu} d\mu' \, \delta\rho(\mu', R_0) \tag{10}$$

where R_0 is the spherical cell radius, and W is the atomic cell volume.

To obtain from Eq. (6) the relation for a shell corrections to the internal energy, one needs the form of the shell density correction. When using the spherically symmetric semiclassical wave function, one can obtain the electron density and particle number expressions:

$$\begin{aligned}\rho(r, \mu) &= \sum_{\{n\}} |\Psi_{\{n\}}(\bar{r})|^2 f\left(\frac{E_{\{n\}} - \mu}{T}\right) \\ &= \frac{1}{2\pi^2} \int d\bar{\kappa} \sum_{n,l} f\left(\frac{E_{nl}(\kappa) - \mu}{T}\right) \frac{2l+1}{r^2 p_{nl}(r)} \frac{dE_{nl}}{dn}\end{aligned} \tag{11}$$

$$N(\mu) = 2 \int d\bar{\kappa} \sum_{n,l} (2l+1) f\left(\frac{E_{nl}(\kappa) - \mu}{T}\right) \tag{12}$$

where κ is quasiimpulse, $p_{nl}(r)=\sqrt{2(E_{nl}-V(r)-(l+1/2)^2/2r^2)}$, and n and l are the principal and angular quantum numbers. If the sums $\sum_{nl}$ in Eqs. (11) and (12) are replaced by integrals $\iint dn\,dl$, all the information about shell effects is lost and we obtain the TF density expression.

The use of the Poisson equation

$$\sum_{n=a}^{b} f_n=\sum_{k=-\infty}^{\infty}\int_{a-\varepsilon}^{b+\varepsilon} dn\, f(n)\cos(2\pi kn), \qquad 0<\varepsilon<1$$

enables us to make the proper substitution $\sum_{nl}\cdots\to\int dn\,dl\cdots$. Subtracting the TF term ($k=s=0$), we obtain the density shell correction and the internal energy shell correction:

$$\delta E_{\text{sh}}=\delta F_{\text{sh}}-T\frac{\delta F_{\text{sh}}}{\partial T}=\int d\bar{r}\left(\frac{3}{2}\rho(r)+\frac{\partial\rho}{\partial\mu}V(r)\right)(\delta\mu-\delta V)+\delta E_2 \tag{13}$$

$$\delta E_2=\frac{1}{2\pi^2}\int d\bar{r}\sum_{k,s}{}'\int d\bar{\kappa}\int dE\,Ef_{\text{F}}\left(\frac{E-\mu}{T}\right)\int dl\,\frac{2l+1}{r^2p_{\text{El}}(r)}\cos(2\pi kn)\cos(2\pi sl) \tag{14}$$

where $\sum_{k,s}'$ denotes the sum without term $k=s=0$.

3. GENERALIZED QUASICLASSICAL QUANTIZATION CONDITION

To calculate the integrals in Eqs. (10)–(14), we need the electron energy spectrum that links E with n and l. To describe the discrete energy levels, one can use the Bohr–Sommerfeld quantization condition

$$S_{\text{El}}=\pi(n-l-1/2) \tag{15}$$

where $S_{\text{El}}=\int dr\,p_{nl}(r)$ is an action. But there are both discrete and band spectra for the compressed atom. The quantization condition, expressed by Eq. (15), is inapplicable in this case. So in order to describe such spectrum features, we need a generalized quantization condition for which a number of requirements must be met:

1. For strongly bound electrons this quantization condition transforms to Eq. (15).
2. For free electron states it describes a continuous spectrum.
3. For $\kappa=0$:

$$\Phi_l'[E_n(\kappa=0),R_0]=0, \qquad l \text{ is even}$$

$$\Phi_l[E_n(\kappa=0),R_0]=0, \qquad l \text{ is odd}$$

where $\Phi_l(E,r)$ is a radial wave function.

4. The function $E(\kappa)$ near the point $\kappa = 0$ is quadratic:

$$E(\kappa) = E(0) + \alpha\kappa^2 + \cdots$$

Let us construct the approximate generalized quantization condition by analogy with the explicit one-dimensional quantization condition (see Ref. 13) and with regard to the above requirements:

$$\tilde{S}_{E1} = \pi(n - l - 1/2) - (-1)^1 \arcsin\left[\tan(\Delta_{E1}) \cos\left(\pi \frac{\kappa}{\kappa_0}\right)\right] \tag{16}$$

where Δ_{E1} characterizes bandwidth

$$\Delta_{E1} = \frac{\pi}{8} + \arctan\left[\tanh\left(\frac{\pi}{4} d_{E1}^2\right)\right], \qquad 0 \leqslant \Delta_{E1} \leqslant \frac{\pi}{4}$$

$$d_{E1}^2 = \frac{p_{E1}^2(R_0) R_0^2}{\beta_1^{1/2}} - \frac{(l + 1/2)^2}{\beta_1^{3/2}}$$

$$\beta_1 = 4\pi\rho(R_0) R_0^4 - 3(l + 1/2)^2 \tag{17}$$

$$\tilde{S}_{E1} = S_{E1} + \frac{d_{E1}^2}{4} \ln \frac{|d_{E1}^2|}{2e} + \frac{1}{2} \arg \Gamma\left(\frac{1 - i d_{E1}^2}{2}\right)$$

$$\arg \Gamma\left(\frac{1 - i d^2}{2}\right) = -\frac{d^2}{2} \Psi\left(\frac{1}{2}\right) - \sum_{n=0}^{\infty} \left[\frac{d^2}{2n + 1} - \arctan\left(\frac{d^2}{2n + 1}\right)\right],$$

$$\Psi\left(\frac{1}{2}\right) = -1.1963510$$

Figure 1 shows a rather good agreement between Eq. (16) and the model [14] results for the electron energy spectrum of the compressed iron.

4. SHELL CORRECTION TO THE PARTICLE NUMBER

Let us calculate the particle number shell correction using Eqs. (16) and (17). When assuming for simplicity the distribution of electron states within the band to be one-dimensional,

$$\int d\bar{\kappa} \cdots = \frac{1}{\kappa_0} \int_0^{\kappa_0} d\kappa \cdots \tag{18}$$

one can obtain the relation for the particle number:

$$N(\mu)=\frac{2}{\pi}\sum_{k}\sum_{l}(2l+1)\int dE\tilde{\tau}_{El}f_F\left(\frac{E-\mu}{T}\right)\cos[2k(\tilde{S}_{El}+\pi(l+1/2))]\,I_k(\varphi_{El})$$

$$\tilde{\tau}_{El}=\frac{\partial\tilde{S}_{El}}{\partial E} \qquad (19)$$

Function $I_k(\varphi_{El})$ is a result of the integration over κ:

$$I_k(\varphi_{El})=\frac{(-1)^k}{2}[P_k(2\varphi_{El}^2-1)-P_{k-1}(2\varphi_{El}^2-1)], \qquad k\neq 0$$

$$I_0(\varphi_{El})=1, \qquad \varphi_{El}=\tan\Delta_{El}$$

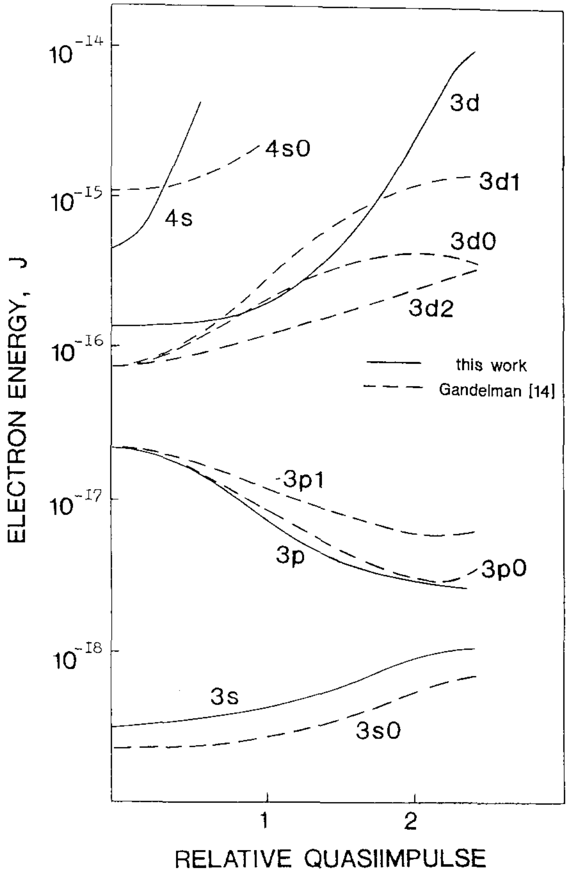

Fig. 1. Electron energy spectrum for strongly compressed iron at temperature $T=0$. Heavy particle density $n_i=3.64\times10^{29}\,\mathrm{m}^{-3}$.

$P_k(X)$ is a Legendre polynomial. For strongly bound electrons $\Delta_{E1} \to 0$, $\varphi_{E1} \to 0$, $I_k(\varphi) \to 1$, and the corresponding energy levels are discrete. So the discrete level contribution to the shell correction to the TF model spectrum is maximal. For sufficiently large energies (free electron states) $\Delta_{E1} \to \pi/4$, $\varphi_{E1} \to 1$, $I_k(\varphi_{E1}) \to 0$, and the corresponding energy state contribution to the shell correction equals zero. Such a spectrum is continuous. There is an intermediate energy region between discrete and continuous spectra in which the shell effect role diminishes. So function $I_k(\varphi_{E1})$ describes shell effect damping when passing from a discrete spectrum to a continuous one.

The atomic spectrum analysis indicates a rather smooth dependence of $E_n(l)$ on l within the shell with the principal quantum number n. But the shells with different n are separated by wide energy intervals (Fig. 2). So in the low-order ξ-expansion series we can replace a summing over discrete l by an integral over l. Then an accurate summing over n by means of the Poisson formula enables us to take into account dominant effects of the atomic shell structure. The term with $k=0$ in Eq. (19) is the TF part. The rest of the sum in Eq. (19) is a shell correction,

$$\delta N_{\rm sh}(\mu) = \frac{2}{\pi} \sum_k{}' \int d\lambda^2 \int dE\, \tilde{\tau}_{E\lambda} f_F\left(\frac{E-\mu}{T}\right) \cos[2k(\tilde{S}_{E\lambda} + \pi\lambda)]\, I_k(\varphi_{E\lambda}) \qquad (20)$$

where $\lambda = l\frac{1}{2}$, and $\sum_k'$ denotes the sum without term $k=0$.

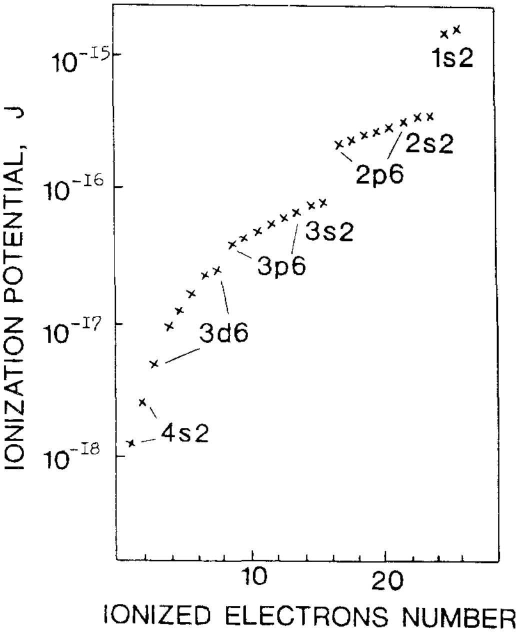

Fig. 2. Ionization potentials for an isolated iron atom.

5. NONDEGENERATE PLASMA

Now let us calculate from Eq. (20) the particle number shell correction for the nondegenerate plasma. The chemical potential μ is negative in this case and $|\mu|/T \gg 1$. So the factor $f_F[(E-\mu)/T]$ in Eq. (20) limits the integration region by discrete spectrum for which $\tilde{S}_{E\lambda} = S_{E\lambda}$, the function $I_k(\varphi_{E1}) \sim 1$ and varies weakly. After integrating by parts over E, one obtains

$$\delta N_{\text{sh}}(\mu) = -\frac{1}{\pi} \sum_k{}' \frac{1}{k} \int dE \frac{\partial f_F}{\partial E} \int d\lambda^2 \sin[2k(S_{E\lambda} + \pi\lambda)]\, I_k(\varphi_{E\lambda}) \qquad (21)$$

To evaluate the integral over λ^2 we use a squared λ-expansion series,

$$S_{E\lambda} = S_E - \pi\lambda - \frac{\delta_E \lambda^2}{2}, \qquad \delta_E = -\left.\frac{\partial^2 S_{E\lambda}}{\partial \lambda^2}\right|_{\lambda=0} \qquad (22)$$

That expansion is obvious for small λ. Figure 3 illustrates the validity of Eq. (22) for any permissible λ $[0 \leqslant \lambda \leqslant \lambda_{\max}(E),\ S_{E\lambda_{\max}(E)} = 0]$ in the treated temperature region. The integral over E is easily calculated because of the derivative $f'_F[(E-\mu)/T]$ behaves like a delta function. Expanding all the

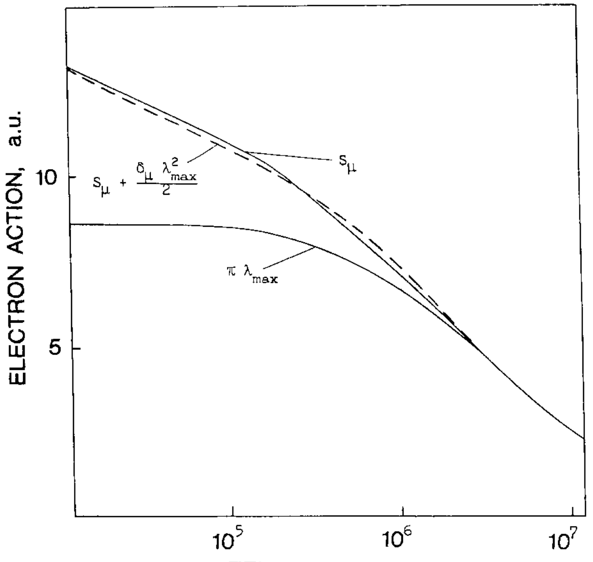

Fig. 3. Approximations for an electron action calculation.

integrand functions in the neighborhood of the point $E=\mu$, one can obtain, finally,

$$\delta N_{\mathrm{sh}}(\mu)=\frac{2}{\pi\delta_{\mu 0}}\sum_{k=1}\left\{\frac{2\pi k\dfrac{\partial\pi\lambda_{\max}}{\partial\mu}T}{\sinh\left(2\pi k\dfrac{\partial\pi\lambda_{\max}}{\partial\mu}T\right)}\cos 2\pi k\lambda_{\max}(\mu)\right.$$

$$\left.-\frac{2\pi k\tau_{\mu 0}T}{\sinh(2\pi\tau_{\mu 0}T)}\cos 2kS_{\mu 0}\right\}\frac{I_k(\varphi_{\mu 0})}{k^2} \tag{23}$$

Equation (23) describes the "temperature" oscillations because of the electron shell ionization with increased temperature. The function $I_k(\varphi_{\mu 0})=1$ for the nondegenerate plasma and tends to zero when the chemical potential becomes positive and increases. Hence, it describes "temperature" oscillation damping when passing from nondegenerate plasma to condensed matter.

A similar computation for the density and energy corrections $\delta\rho_{\mathrm{sh}}$ and δE_2 gives the next relation:

$$\delta E_2=\mu\int d\bar{r}\,\delta\rho_{\mathrm{sh}}=\mu\,\delta N_{\mathrm{sh}}(\mu) \tag{24}$$

Qualitative evaluation and numerical calculations show that the shell correction δV_{sh} on the average is significantly smaller than the chemical potential correction $\delta\mu_{\mathrm{sh}}$. So $\delta\mu_{\mathrm{sh}}$ may be calculated approximately from the normalization condition

$$\delta\mu_{\mathrm{sh}}=-\delta N_{\mathrm{sh}}\Big/\int d\bar{r}\frac{\partial\rho}{\partial\mu} \tag{25}$$

and the internal energy correction δE_{sh} may be determined from Eqs. (13), (24), and (25),

$$\delta E_{\mathrm{sh}}=\left\{\frac{3}{2}Z-\int d\bar{r}\frac{\partial\rho}{\partial\mu}\left[\mu-V(r)\right]\right\}\delta\mu_{\mathrm{sh}} \tag{26}$$

Analysis of the pressure correction, expressed by Eq. (10), discloses that the second term is exponentially small for the nondegenerate plasma. The reason is that the cell boundary region is classically forbidden for the electron states with negative energies. So for the nondegenerate plasma, one can use

$$\delta P_{\mathrm{sh}}=\rho(R_0)\,\delta\mu_{\mathrm{sh}} \tag{27}$$

Equations (23) and (25)–(27) allow us to calculate the shell corrections to the nondegenerate plasma equation of state. Figures 4–7 show the

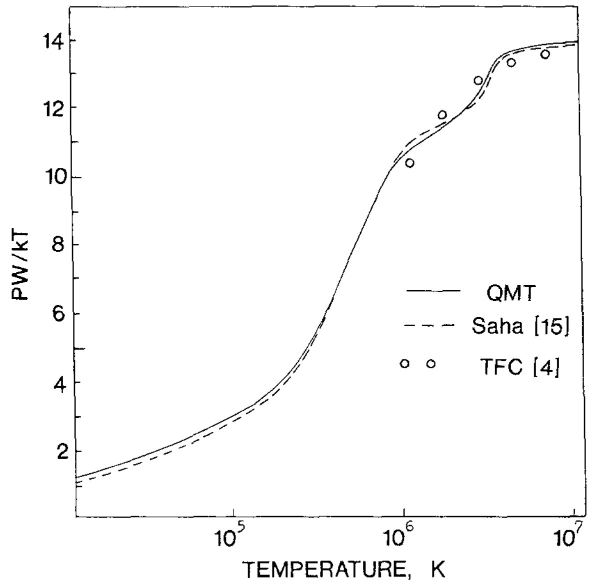

Fig. 4. Thermal equation of state of aluminum. Isochore for heavy particle density $n_i = 10^{27}\ \mathrm{m}^{-3}$.

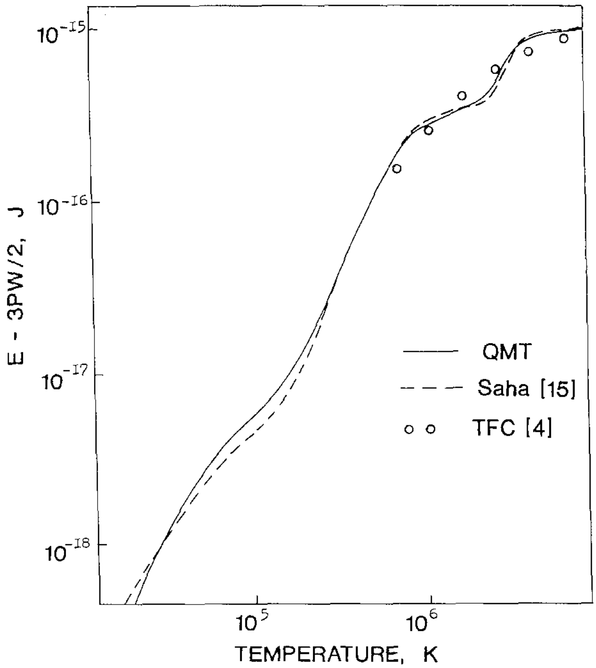

Fig. 5. Calorific equation of state of aluminum. Isochore for heavy particle density $n_i = 10^{27}\ \mathrm{m}^{-3}$.

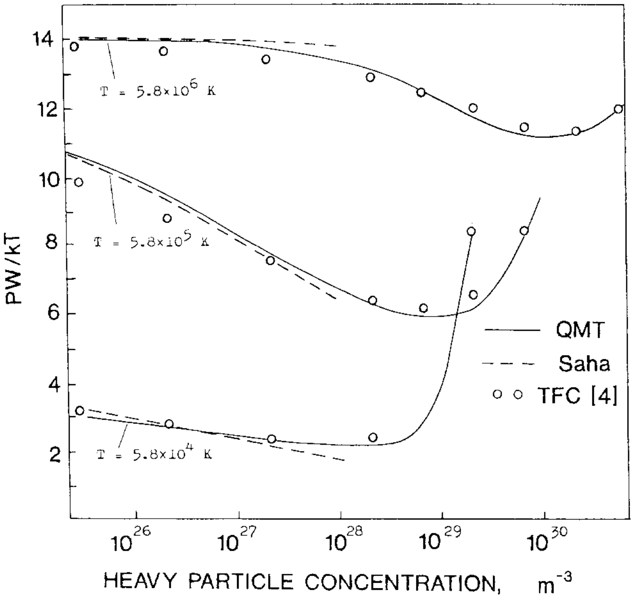

Fig. 6. Thermal equation of state of aluminum. Isochore for different temperatures.

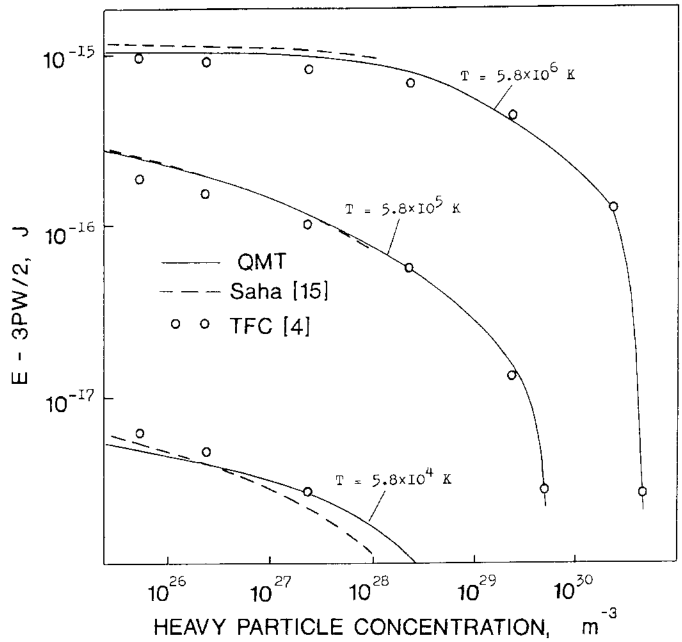

Fig. 7. Calorific equation of state of aluminum. Isochore for different temperatures.

comparison of the suggested model results with both the plasma [15] and the statistical [4] model calculations. Considerable deviation of the statistical model confirms the important shell effects role in plasma region as noted in Ref. 16.

6. QUASICLASSICAL TEMPERATURE MODEL (QMT)

An extension of Eqs. (23) and (25)–(27) over all ranges of the parameters with Eqs. (4), (5), (8), and (9) enables us to construct the quasiclassical "temperature" model (QMT), which turns into the TFK model for the degenerate matter and checks well with Saha model for the Boltzmann plasma. Indeed all the "temperature" shell corrections vanish for the degenerate matter because of the factor $I_k(\varphi_{\mu 0})$. Of course in the intervening region of the parameters QMT is a reasonable physical interpolation only. Thus, one can use this model instead of the mathematical union of the TFK and Saha models [17] without any joining problems or any empirical data. The other great virtue of the QMT is its Z-scaling: a single set of calculations for one Z suffices for all Z. This virtue is known

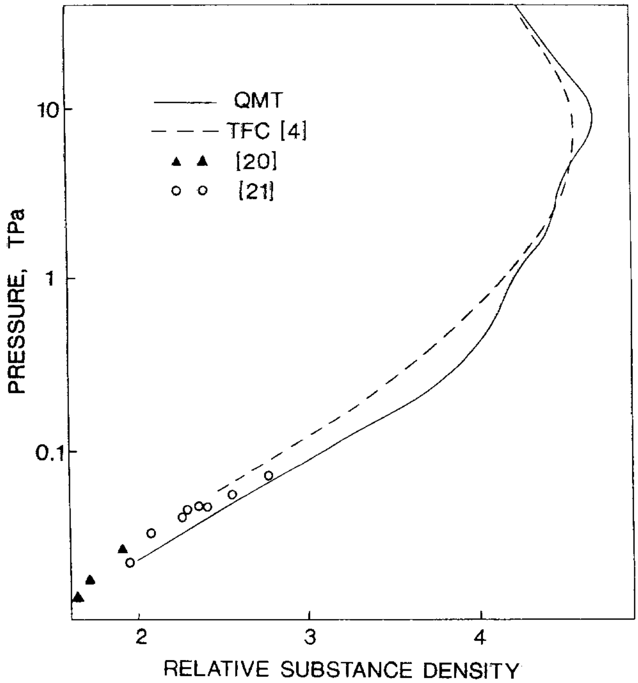

Fig. 8. Shock Hugoniot for lithium. Symbols are experimental measurements.

for TF and TFK models but one can use it for the shell correction too. Indeed let us separate the Z-dependence of all the quantities in Eqs. (23)–(26):

$$S_\mu^{(Z)} = S_\mu^{(1)} Z^{1/3}, \qquad \tau_\mu^{(Z)} = \tau_\mu^{(1)}/Z, \qquad \pi\lambda_{\max}^{(Z)} = \pi\lambda_{\max}^{(1)} Z^{1/3},$$

$$\delta_\mu^{(Z)} = \delta_\mu^{(1)}/Z^{1/3}, \qquad \left(\frac{\partial\pi\lambda_{\max}}{\partial\mu}\right)^{(Z)} = \left(\frac{\partial\pi\lambda_{\max}}{\partial\mu}\right)^{(1)} \Big/ Z$$

$$\left(\int d\bar{r}\,\frac{\partial\rho_{TF}}{\partial\mu}\right)^{(Z)} = \left(\int d\bar{r}\,\frac{\partial\rho_{TF}}{\partial\mu}\right)^{(1)} \Big/ Z^{1/3}$$

$$\left\{\int d\bar{r}\,\frac{\partial\rho_{TF}}{\partial\mu}\left[\mu_{TF} - U_{TF}(r)\right]\right\}^{(Z)} = \left\{\int d\bar{r}\,\frac{\partial\rho_{TF}}{\partial\mu}\left[\mu_{TF} - U_{TF}(r)\right]\right\}^{(1)} Z$$

The procedure for Z-scaling calculation of the shell thermodynamic terms and needed tables are assumed in Ref. 18.

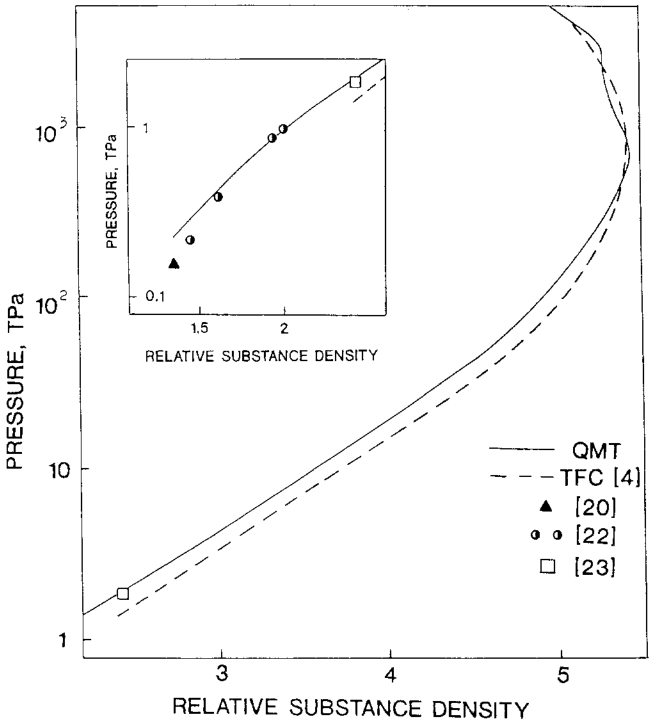

Fig. 9. Shock Hugoniot for molybdenum. Symbols are experimental measurements.

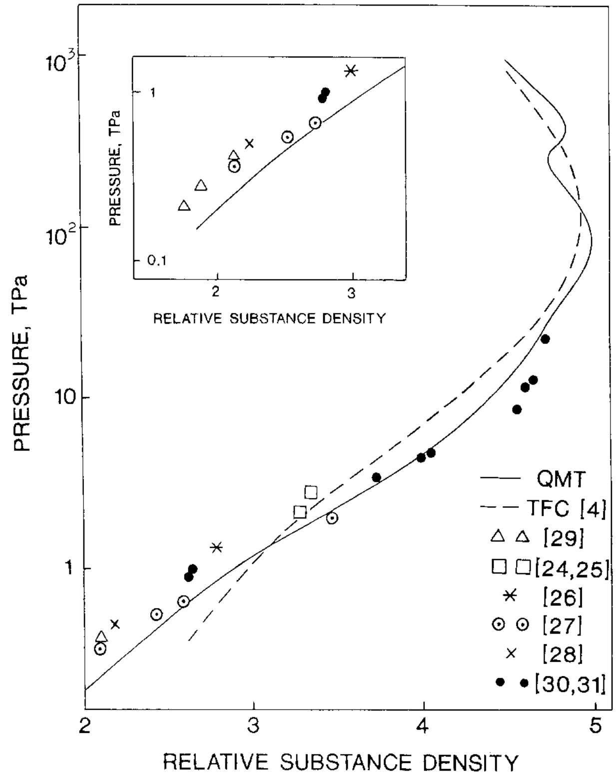

Fig. 10. Shock Hogoniot for aluminum. Symbols are experimental measurements.

For demonstration QMT Hugoniot curves of some metals are shown in Figs. 8–10 with the TFK-model results and the available experimental data. We used in our calculations the one-component plasma theory [19] for the nuclear motion term.

7. CONCLUSIONS

A simple quasiclassical Z-scaling model is proposed to calculate the electron equation of state for the matter with a higher energy concentration. This model may be employed over a wide range of densities and temperatures from the Saha model region of application to the improved Thomas–Fermi model (TFK) area of use. The proposed model describes ab initio typical step behavior of the ionization state and energy as a result of successive shell ionization with increased temperature. The model naturally includes the effects of electron–ion interaction with increased density.

ACKNOWLEDGMENT

The authors thank Professor V. E. Fortov for fruitful discussions.

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