Convergence properties as a function of spatial dimensionality of gradient expansions for the ground-state energy of an inhomogeneous electron gas

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The extended Thomas-Fermi approximation for the ground-state energy of a many-fermion system is generalized to arbitrary spatial dimension. Our objective is a better understanding of convergence properties of such gradient expansions with a view to applications to systems of reduced dimensionality or esoteric geometry. The convergence is tested and found to be adequate by comparing to an exact result for the surface kinetic energy of a semi-infinite system. Both local and nonlocal contributions to the exchange energy are also given for arbitrary dimension. The extension to the thermodynamic free energy at finite temperature for arbitrary dimension is also discussed.

There have been several interesting studies recently of intrinsically inhomogeneous many-fermion systems which exhibit novel ground states (and presumably complete phase diagrams) which are related to the fact that their effective spatial dimensionality is less than three. Typically, this reduction in dimensionality is a consequence of strong externally applied fields of competing internal interactions or geometrical constraints. Examples would include an electron gas in very intense magnetic fields (which exhibits a filamentary crystalline structure with implications for the crust of neutron stars[1]), dense nuclear matter just below the nuclear saturation density[2,3] (which exhibits a bubble phase as well as phases of different geometry[2]), various quasi-two-dimensional electronic devices with microstructures (which exhibit a wide variety of new effects and have rich applications), and many others.[4]

For the most part, these systems have been studied theoretically by rather simple approximations which served to expose the esoteric features. There is a need for a systematic and more accurate treatment which, at the same time, is sufficiently simple for further exploratory work. For three-dimensional systems, it is well known that a very useful tool has been provided by the Thomas-Fermi (TF) approximation or an extended Thomas-Fermi (ETF) theory in which nonlocality is incorporated in energy functionals by means of gradient corrections.[5-10] It is natural to consider ETF in systems of reduced dimensionality as well. To our knowledge, this has not previously been available but should be very useful.

In this paper, we give explicit results for the ETF ground-state energy functional for a many-fermion system in which the spatial dimensionality d is an arbitrary continuous parameter. We obtain the difficult nonlocal contributions through fourth order in powers of density gradients for the kinetic energy. This is known to be the appropriate level of approximation for three-dimensional systems, both for electronic systems[11-13] and for nuclear applications,[14] when parametrized density functions are used. We discuss interparticle two-body-interaction contributions to the local-density approximation (LDA) and to gradient corrections to leading order in powers of the interaction and to second order in power of density gradients. We then study the convergence properties of the gradient-expansion series as a function of spatial dimensionality and find it to be satisfactory. This is an extremely important point.

The starting point in our calculation is the classical partition function $Z(\beta)$ for noninteracting fermions in the presence of an external single-particle potential $V(\mathbf{r})$. The expansion for $Z(\beta)$ giving systematic corrections to the TF results is the Wigner-Kirkwood expansion.[5] The parameter β is to be treated as a variable in calculating the free energy E at zero temperature. Expressing E in terms of $Z(\beta)$ we obtain to fourth order in gradients,

$$E=\mu N-2\mathcal{L}_{\mu}^{-1}\left[\frac{Z(\beta)}{\beta^2}\right]\approx\mu N-\left[\frac{2m}{\hbar^2}\right]^{d/2}\frac{1}{2^{d-1}\pi^{d/2}}$$
$$\times\int d^dr\left\{\frac{(\mu-V)^{1+d/2}}{\Gamma(2+d/2)}-\frac{1}{12}\left[\frac{\hbar^2}{2m}\right]\frac{(\mu-V)^{d/2-1}}{\Gamma(d/2)}\nabla^2V\right.$$
$$\left.+\frac{1}{1440}\left[\frac{\hbar^2}{2m}\right]^2\frac{(\mu-V)^{d/2-2}}{\Gamma(d/2-1)}\left[-7\nabla^4V+5\nabla\cdot\left[\frac{(\nabla^2V)^2\nabla V}{(\nabla V)^2}\right]+\nabla\cdot\left[\frac{\nabla^2(\nabla V)^2\nabla V}{(\nabla V)^2}\right]\right]\right\}. \tag{1}$$

Here $\mathcal{L}_{\mu}^{-1}$ denotes an inverse Laplace transform with respect to the chemical potential μ. A gradient expansion for the electron number N in gradients of V may also be obtained with the use of $N=2\mathcal{L}_{\mu}^{-1}[Z(\beta)/\beta]$. From Eq. (1), we obtain an expansion for the grand canonical potential at zero temperature

$$\Omega=E-\mu N=\Omega_{\mathrm{LDA}}+\Omega_2+\Omega_4+\cdots .$$

Expressed in terms of the TF density $n_{\rm TF}(\mathbf{r})$ defined by

$$\mu - V(\mathbf{r}) \equiv (\hbar^2/2m)[n_{\rm TF}(\mathbf{r})d/2K_d]^{2/d} ,$$

where $K_d = \Omega_d/(2\pi)^d$ and $\Omega_d = 2\pi^{d/2}/\Gamma(d/2)$, we have

$$\Omega_{\rm LDA}[n_{\rm TF}] = -\frac{\hbar^2}{2m}\left(\frac{d}{2K_d}\right)^{2/d}\left(\frac{2}{d+2}\right)\int d^dr\, n_{\rm TF}^{1+2/d} , \tag{2a}$$

$$\Omega_2[n_{\rm TF}] = \frac{\hbar^2}{2m}\left(\frac{d-2}{12d}\right)\int d^dr \frac{(\nabla n_{\rm TF})^2}{n_{\rm TF}} , \tag{2b}$$

$$\Omega_4[n_{\rm TF}] = \frac{\hbar^2}{2m}\left(\frac{2K_d}{d}\right)^{2/d}\frac{1}{2880d}(d-2)(4-d)\int d^dr\, n_{\rm TF}^{-1-2/d}\left[12(\nabla^2 n_{\rm TF})^2 - 16\frac{\nabla^2 n_{\rm TF}(\nabla n_{\rm TF})^2}{n_{\rm TF}} + \left(\frac{2+5d}{d}\right)\frac{(\nabla n_{\rm TF})^4}{n_{\rm TF}^2}\right] . \tag{2c}$$

For noninteracting systems, the kinetic energy is given by $T = \Omega + \int d^dr(\mu - V)n(\mathbf{r})$, where the exact number density is given by $n(\mathbf{r}) = n_{\rm TF} + \delta n_g$, with $\delta n_g = \delta_2 n + \delta_4 n + \cdots$, and the subscripts 2, 4, ... correspond to the order of the gradient correction. Equating terms of the same order in gradients, we obtain a formal expansion for the kinetic energy,

$$T[n] = T_{\rm LDA}[n] + T_2[n] + T_4[n] + \cdots , \tag{3}$$

where

$$T_{\rm LDA}[n] = \frac{\hbar^2}{2m}\left(\frac{d}{2K_d}\right)^{2/d}\left(\frac{d}{d+2}\right)\int d^dr\, n^{1+2/d} , \tag{4a}$$

$$T_2[n] = \frac{\hbar^2}{2m}\left(\frac{d-2}{12d}\right)\int d^dr \frac{(\nabla n)^2}{n} , \tag{4b}$$

$$T_4[n] = \frac{\hbar^2}{2m}\left(\frac{2K_d}{d}\right)^{2/d}\left(\frac{d-2}{720d}\right)\int d^dr\, n^{-1-2/d}\left[2(d+1)(\nabla^2 n)^2 - (d+6)\frac{\nabla^2 n(\nabla n)^2}{n} + \frac{2}{d}(d+1)\frac{(\nabla n)^4}{n^2}\right] . \tag{4c}$$

In order to test the convergence of this gradient expansion for the kinetic-energy functional, exact results are required for a standard reference system. The surface energy of a bounded semi-infinite many-fermion system provides a suitable test case of physical interest for both electronic[11,15–17] and nuclear[18] problems. We thus consider a plasma bounded by a $(d-1)$-dimensional hyperplane perpendicular to the z direction. Equation (4) gives the gradient expansion for the surface kinetic energy per unit area $\sigma_s = \sigma_{s0} + \sigma_{s2} + \sigma_{s4} + \cdots$. Assuming that the plasma is constrained to be in the region $z \geq 0$ by a potential barrier of height $V_0 = \hbar^2 k_0^2/2m$ when $z \leq 0$, the electron density is, for this finite-barrier model (FBM),

$$n(z) = C_d\int_0^{k_F} dk_z (k_F^2 - k_z^2)^{(d-1)/2}[\sin^2(k_z z) + (k_z/k_0)^2\cos(2k_z z) + (k_z\kappa/k_0^2)\sin(2k_z z)], \quad z > 0 , \tag{5a}$$

$$= C_d\int_0^{k_F} dk_z (k_F^2 - k_z^2)^{(d-1)/2}(k_z/k_0)^2 e^{2\kappa z}, \quad z < 0 , \tag{5b}$$

where k_F is the Fermi wave number, $\kappa \equiv (k_0^2 - k_z^2)^{1/2}$ and $C_d \equiv 2^{4-d}/[\pi^{(d+1)/2}(d-1)\Gamma((d-1)/2)]$. The contributions to σ_s for the FBM are given in Fig. 1, where the exact results for the surface kinetic energy are also given. In a straightforward way, we obtain the exact surface kinetic energy for the FBM by applying the Euler-Maclaurin series to the kinetic energy summed over the eigenstates as $\sigma_s = \sigma_s(1) + \sigma_s(2)$, where

$$\sigma_s(1) = \frac{\hbar^2}{2m}k_F^{d+1}\frac{K_{d-1}}{(d-1)(d+1)(d+2)} , \tag{6a}$$

$$\sigma_s(2) = \frac{\hbar^2}{m}k_0^{d+1}\frac{K_{d-1}}{\pi(d+1)}\left(\phi_{d+1}(k_F/k_0) - \frac{d(d+1)}{(d-1)(d+2)}(k_F/k_0)^2\phi_{d-1}(k_F/k_0)\right) , \tag{6b}$$

where we have introduced the integral

$$\phi_d(x) \equiv \int_0^{\sin^{-1}x} dt\,(x^2 - \sin^2 t)^{d/2} . \tag{7}$$

$\sigma_s(1)$ is the surface kinetic energy for the infinite-barrier model and $\sigma_s(2)$ gives the correction for the FBM.[15–17]

The numerical results in Fig. 1 show that the surface kinetic energy with fourth-order gradient corrections improves as the dimensionality increases. At the same time, the LDA σ_{s0} gets increasingly poor in the limit of large dimensionality. The value of the electron density for $z >> k_F^{-1}$ is $n_B = (2K_d/d)k_F^d$. The kinetic energy at different densities scales as r_s^{-d-1}, where $r_s \equiv [d\Gamma(d/2)/2n_B a_0^d \pi^{d/2}]^{1/d}$ and a_0 is the Bohr radius. The gradient expansion up to fourth order converges very well towards σ_s for barrier heights $1 < \mu/V_0 \lesssim 2$. As the barrier height increases, the convergence gets poorer and for the infinite-barrier model σ_{s4} diverges. This behavior has been confirmed for the range $2 < d < 30$ and is assumed to be a general feature of the square-barrier model of arbitrary dimensionality. The $d = 3$ results are given in Ref. 11.

An enlightening model calculation can be based on the spatial density profile $n(z) = n_B/[1 + \exp(-\alpha z)]$. For this model, the integrals involved are elementary and we obtain

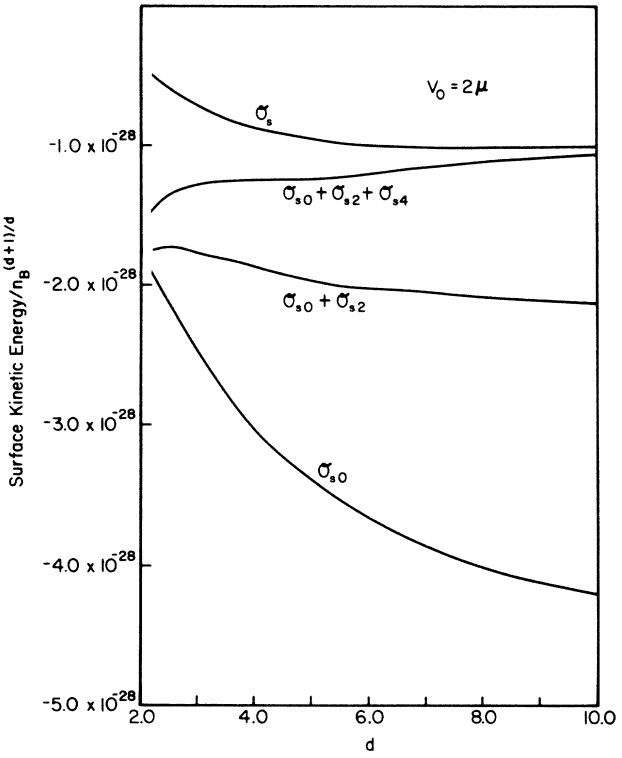

FIG. 1. Plot of the surface kinetic energy for the finite-barrier model as a function of d. The barrier height is chosen as $V_0=2\mu$. The *exact* value σ_s, given by Eqs. (6), is compared with the gradient expansion. σ_{s0} is the local-density approximation and σ_{s2}, σ_{s4} are the gradient corrections to second and fourth order, respectively, n_B is the bulk density and cgs units are used. Note the improvement in convergence as d increases.

the contributions to σ_s in the gradient expansion as

$$\sigma_{s0}=-\frac{\hbar^2}{2m}\left(\frac{d}{2K_d}\right)^{2/d}\left(\frac{d}{d+2}\right)\frac{n_B^{1+2/d}}{\alpha}\left[\psi\left(1+\frac{2}{d}\right)+\gamma_E\right], \quad (8a)$$

$$\sigma_{s2}=\frac{\hbar^2}{2m}\left(\frac{d-2}{24d}\right)n_B\alpha, \quad (8b)$$

$$\sigma_{s4}=\frac{\hbar^2}{2m}\left(\frac{2K_d}{d}\right)^{2/d}\frac{1}{720}n_B^{1-2/d}\alpha^3\frac{d(d+1)(d^2-2d+2)}{(2d-1)(3d-2)(d-1)}, \quad (8c)$$

where ψ and γ_E are the digamma function and Euler's constant, respectively. An interesting aspect of Eqs. (8) is that the threatened disappearance of the fourth-order gradient term at $d=2$ in Eq. (4c) is canceled by an infinite value of the integral in σ_{s4}, resulting in a *finite* contribution for a two-dimensional system. It can be verified that this is a consequence of the exponential decay of the particle density in the vacuum region. Clearly, this feature is general and is not limited to the above model density profile. There are no published results for the gradient corrections in two dimensions for the kinetic energy. Our results show that the gradient corrections to the TF result must be handled delicately. The numerical results based on Eqs. (8) are qualitatively similar to those given in Fig. 1, when $\alpha \gtrsim 2k_F$. There are of course no *exact* results for σ_s corresponding to the model density which yields Eqs. (8).

The conclusions that the gradient expansion converges well for systems of large dimensionality and that the LDA is inappropriate in this limit are based on exact calculations within the FBM. Calculations based on Eqs. (8) also show that, like the FBM, the gradient corrections become more significant as d increases. As a consequence, even though the LDA becomes poorer as d increases, the behavior of the gradient corrections truncated at fourth order is *precisely* what is required to obtain very good agreement with the exact results for all d.

We now turn to the effects of the interparticle interaction.[19] We give the exchange energy functional for the Coulomb potential $e^2r^{2-d}/(d-2)$. The LDA for the exchange energy and the first-order gradient correction are

$$E_x^{\mathrm{LDA}}[n]=-\frac{1}{8}Q_d\left(\frac{2K_d}{d}\right)^{2/d}\frac{d^2}{(d-1)(d-2)}\int d^dr n^{2(d-1)/d}, \quad (9)$$

$$E_x^{(2)}[n]=\frac{Q_d}{192d}(3d^2-16d+28)\left(\frac{2K_d}{d}\right)^{4/d}\int d^dr\frac{(\nabla n)^2}{n^{4/d}}, \quad (10)$$

where $Q_d\equiv e^2\Omega_d$. The factor $(d-2)^{-1}$ in Eq. (9) is due to the choice of interaction potential and should be contrasted to the LDA for E_x obtained by Glasser and Boersma[20] who used the $1/r$ potential in all dimensions. Substituting the model density profile used for obtaining Eqs. (8) into Eqs. (9) and (10), we obtain the surface exchange energy per unit area as

$$E_x^{s0}=\frac{1}{8\alpha}Q_d\left(\frac{2K_d}{d}\right)^{2/d}\frac{d^2}{(d-1)(d-2)}n_B^{2-2/d}\left[\psi\left(2-\frac{2}{d}\right)+\gamma_E\right], \quad (11)$$

$$E_x^{s2}=-\frac{\alpha d}{384}Q_d\frac{(3d^2-16d+28)}{(2-d)(4-3d)}\left(\frac{2K_d}{d}\right)^{4/d}n_B^{2-4/d}. \quad (12)$$

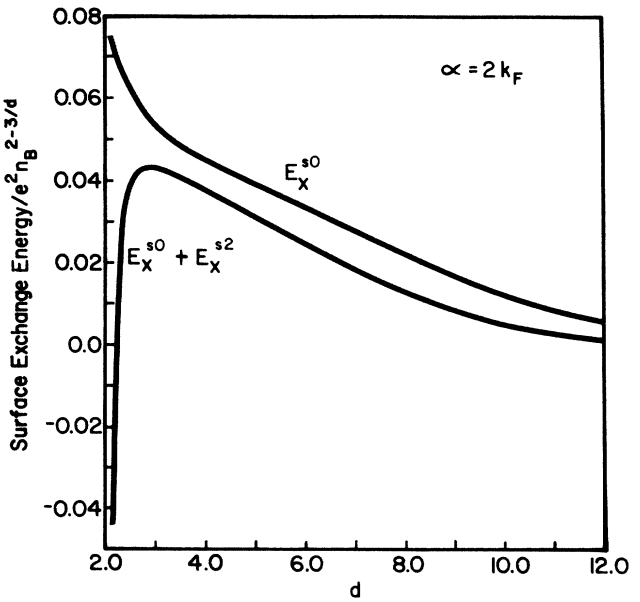

FIG. 2. Surface exchange energy given by Eqs. (11) and (12), based on the model profile $n(z)=n_B/[1+\exp(-\alpha z)]$ described in the text. cgs units are used.

In Fig. 2, the surface exchange energy given by Eqs. (11) and (12) is plotted as a function of d. Note that the gradient corrections to the LDA especially for d near 2 are now dependent on the precise details of the assumed interaction as well as on the density profile.[21]

Finally, in order to construct full phase diagrams, we require the free energy $F=\Omega+\mu N$ at finite temperature. The calculations are lengthy and, for brevity, we give only the LDA and the lowest-order gradient correction as functionals of the exact density.

$$F[n]=\int d^d r\left[V(\mathbf{r})n(\mathbf{r})+k_B T\left(\frac{2mk_B T}{\hbar^2}\right)^{d/2} K_d\left[\eta(\mathbf{r})I_{d/2-1}(\eta(\mathbf{r}))-\frac{2}{d}I_{d/2}(\eta(\mathbf{r}))\right]\right.$$

$$\left.+\frac{\hbar^2}{24m}\left(\frac{d-2}{d-1}\right)\left(\frac{\hbar^2}{2mk_B T}\right)^{d/2}\frac{I_{d/2-3}(\eta(\mathbf{r}))}{K_d I^2_{d/2-2}(\eta(\mathbf{r}))}(\nabla n(r))^2\right] .$$

The local parameter $\eta(\mathbf{r})=\mu_0(n(\mathbf{r}))/k_B T$ is obtained from

$$n(\mathbf{r})=(2mk_B T/\hbar^2)^{d/2}K_d I_{d/2-1}(\eta(\mathbf{r})) ,$$

where the standard Fermi-Dirac integrals are defined by

$$I_\lambda(\eta)=\int_0^\infty dx\frac{x^\lambda}{1+\exp(x-\eta)}$$

for $\lambda>-1$ and otherwise by the recurrence relation $I_\lambda(\eta)=(\lambda+1)^{-1}I'(\eta)$, but for the limiting case of $\lambda=-1$.

Further corrections due to higher-order gradients and to interparticle interactions as well as applications will be given elsewhere.

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