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Exact hierarchy of equations for the kinetic energy functional

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The virial theorem is used to derive an exact hierarchy of coupled equations for the kinetic energy functional $T[\rho]$ of an inhomogeneous system of noninteracting fermions. The hierarchy links the nth functional derivative of $T[\rho]$ with respect to the particle density $\rho(\mathbf{r})$ to the $(n+1)$th functional derivative and to $\rho(\mathbf{r})$ for $n=1,2,\ldots$. These functional derivatives are the analogs of the so-called "direct correlation functions" in the theory of classical fluids, and the hierarchy obtained resembles the Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy for the distribution functions associated with such fluids. Some general properties of the two- and three-point correlation functions are described, in particular, their relations to the linear and quadratic response functions that describe the response of the inhomogeneous system to a change in the potential. The hierarchy is then used to obtain several equivalent expressions for $T[\rho]$, one of which is an infinite series of successively higher-order terms, of which the nth term represents the contribution from the n-point correlation function and the densities associated with those points. The equations are then examined in the homogeneous (uniform) gas limit and the first and second equations are verified explicitly with the aid of the associated response functions. The analysis indicates that the infinite series expression converges rapidly for the three-dimensional gas; it also reveals that for both the one-dimensional and two-dimensional gas, the series terminates after the first few terms, suggesting that the structure of $T[\rho]$ is much simpler in one and two dimensions than in three dimensions.

I. INTRODUCTION

A quantity that plays a fundamental role in density-functional theory is the kinetic energy $T[\rho]$ considered as a functional of the particle density $\rho(\mathbf{r})$. A knowledge of the functional $T[\rho]$, or accurate approximations to it, makes possible the calculation of ground-state properties of many-fermion systems without the calculation of the associated wave functions. In the years following the birth of quantum mechanics, Thomas[1] and Fermi[2] made the first approximation, in which $T[\rho]$ is a simple, algebraic function of the local density $\rho(\mathbf{r})$. Their ideas were soon followed by von Weizsäcker[3] with the addition of a term containing the square of the density gradient. The resulting Thomas–Fermi–von Weizsäcker (TFW) approximation, and others that followed through inclusion of higher-order gradient corrections, give $T[\rho]$ in terms of the local density and its spatial derivatives.[4]

Since the seminal work of Hohenberg and Kohn,[5] which has provided a foundation for much of the work in density-functional theory in the past 25 years, it has been recognized that nonlocal contributions to $T[\rho]$ play an important role, for example when attempting to describe some features of inhomogeneous systems such as the oscillations of the electron density in an atom (which reflect the shell structure). Herring[6] has recently presented a thorough survey of the field and summarized in detail the relevant concepts and relationships which enter into calculations of $T[\rho]$.[7]

The purpose of this paper is to derive and discuss a hierarchy of coupled, exact equations which link the kinetic energy functional $T[\rho]$ of an inhomogeneous system of noninteracting fermions to a set of correlation functions which are the analogues of the so-called "direct correlation" functions in the theory of classical fluids.[8] More precisely, the hierarchy links the functional derivatives of $T[\rho]$ with respect to the density at different points in the system. The equations bear a certain resemblance to the classical Bogoliubov-Born-Green Kirkwood-Yvon (BBGKY) hierarchy[9] which relates the nth order distribution function to the $(n+1)$th-order distribution function and the interaction potential.[10]

The basis of the present derivation is provided by the virial theorem of quantum mechanics[11] which relates the kinetic energy of the particles to the density $\rho(\mathbf{r})$ and the gradient of the potential, $\nabla u(\mathbf{r})$. Our starting point is the version of the virial theorem as given by Bartolotti and Parr,[12] Ghosh and Parr,[13] Levy and Perdew,[14] and Herring,[6] in which reference to the potential $u(\mathbf{r})$ has been eliminated in favor of an expression involving $\rho(\mathbf{r})$.

The paper is organized as follows. Section II gives the familiar, basic relations on which the subsequent equations and theory are built. In Sec. III we derive the hierarchy of exact equations which determine the structure of $T[\rho]$ for any inhomogeneous, noninteracting system of fermions. These equations allow us to express $T[\rho]$ in several equivalent forms; the latter are presented in Sec. IV. In Sec. V we give the general relations which link the correlation functions entering into the hierarchy to the more familiar response functions which describe the linear, quadratic, and higher-order response of the inhomogeneous system to a perturbation $\Delta u(\mathbf{r})$ of the potential. Section V also points out some general consequences which follow from the assumption of stability of the system. In Sec. VI we test the validity of the hierarchy by considering the homogeneous (uniform) Fermi gas for which the linear-response function is well known[15] and the quadratic response function can be calculated. In this way, the first and second equations of the hierarchy are explicitly verified for both the one-dimensional and the three-dimensional gas. The analysis also suggests that for the one-dimensional and two-dimensional gas, the functional $T[\rho]$ must have a much simpler structure than for the three-dimensional gas. Some concluding remarks are given in Sec. VII. In Appendix A, the connection between the correlation functions used in this work and the N-particle response functions calculable from steady-state perturbation theory is presented in detail and a number of useful, general identities and relationships are established. Finally, in Appendix B, these identities are considered in the homogeneous gas case where they assume a much simpler and more transparent form.

II. BASIC RELATIONS

We consider a system of N noninteracting fermions of mass m in a static, spin-independent potential $u(\mathbf{r})$. The Hamiltonian consists of the kinetic energy operator $\hat{T}$ and potential energy operator $\hat{U}$:

$$\hat{H}=\hat{T}+\hat{U}=\sum_{i=1}^{N}\frac{\mathbf{p}_i^2}{2m}+\int d\mathbf{r}\hat{\rho}(\mathbf{r})u(\mathbf{r}) \ , \tag{1}$$

where $\hat{\rho}(\mathbf{r})=\sum_{i=1}^{N}\delta(\mathbf{r}-\mathbf{r}_i)$ is the particle density operator. The ground state $|\Phi_0\rangle$, the associated density $\rho(\mathbf{r})$, and the ground-state energy E_N are functionals of $u(\mathbf{r})$ and are given by the expressions

$$E_N[u]=\langle\Phi_0|\hat{H}|\Phi_0\rangle=\langle\Phi_0|\hat{T}|\Phi_0\rangle+\int d\mathbf{r}\,\rho(\mathbf{r})u(\mathbf{r}) \ , \tag{2}$$

$$\rho(\mathbf{r})=\langle\Phi_0|\hat{\rho}(\mathbf{r})|\Phi_0\rangle, \quad \int d\mathbf{r}\rho(\mathbf{r})=N \ . \tag{3}$$

$|\Phi_0\rangle$ is an N-particle determinant of one-particle states φ_i with associated energy level ε_i. Hence E_N and $\rho(\mathbf{r})$ are given, respectively, by the sum of the energy eigenvalues ε_i and by the sum of one-particle probability densities $|\varphi_i(\mathbf{r})|^2$ associated with the N lowest levels. It is worth noting that $\rho(\mathbf{r})$ is also given formally by the functional derivative of $E_N[u]$ with respect to $u(\mathbf{r})$ at constant particle number N:

$$\rho(\mathbf{r})=\left.\frac{\delta E_N[u]}{\delta u(\mathbf{r})}\right|_N \ . \tag{4}$$

The virial theorem of quantum mechanics[11] relates the expectation value of the kinetic energy, $\langle\hat{T}\rangle$, to the particle density $\rho(\mathbf{r})$ and the gradient of the potential $\nabla u(\mathbf{r})$:

$$\langle\hat{T}\rangle\equiv\langle\Phi_0|\hat{T}|\Phi_0\rangle=\tfrac{1}{2}\int d\mathbf{r}\rho(\mathbf{r})\mathbf{r}\cdot\nabla u(\mathbf{r}) \ . \tag{5}$$

For example, for the quadratic (harmonic oscillator) potential, Eq. (5) gives $\langle\hat{T}\rangle=\langle\hat{U}\rangle$, while for noninteracting electrons in an attractive Coulomb potential, Eq. (5) leads to $\langle\hat{T}\rangle=-\langle\hat{U}\rangle/2$. It is important to note that Eq. (5) holds also when $u(\mathbf{r})$ vanishes inside a finite volume V containing the particles; in that case, near the surface the quantity $\rho(\mathbf{r})\nabla u(\mathbf{r})$ takes into account the pressure gradient of the forces acting at the surface, and the right-hand side of (5) then gives the quantity $pVd/2$, where p is the pressure applied at the surface and d is the dimensionality of the system.[16]

A quantity which plays an important role in this paper is the chemical potential $\mu[u]$ which is defined as the derivative

$$\mu[u]=\left.\frac{\partial E_N[u]}{\partial N}\right|_u \ . \tag{6}$$

Here the notation indicates that the derivative must be taken with $u(\mathbf{r})$ held constant. This convention will be followed throughout.

Density-functional theory provides an alternative approach to the calculation of ground-state properties of the system. This approach starts from the fact that the kinetic energy of the system is a unique functional $T[\rho]$ of the particle density. In the density-functional approach, the ground-state properties are determined by a variational procedure in which the density is allowed to vary at fixed potential $u(\mathbf{r})$ and chemical potential μ. For this purpose one introduces the quantity Ω defined as

$$\Omega=E-\mu N=T[\rho]+\int d\mathbf{r}\rho(\mathbf{r})[u(\mathbf{r})-\mu] \ , \tag{7}$$

where μ is to be considered a Lagrange multiplier. The equilibrium density is determined by minimizing Ω with respect to arbitrary variations in $\rho(\mathbf{r})$ at constant $u(\mathbf{r})$ and μ. $\rho(\mathbf{r})$ at the minimum is the ground-state density; the equation determining the minimum is

$$\left.\frac{\delta\Omega}{\delta\rho(\mathbf{r})}\right|_{u,\mu}=\left.\frac{\delta T[\rho]}{\delta\rho(\mathbf{r})}\right|_{u,\mu}+u(\mathbf{r})-\mu=0 \ . \tag{8}$$

This functional equation will be rewritten as

$$f(\mathbf{r};[\rho])=\mu-u(\mathbf{r})\equiv-\bar{u}(\mathbf{r}) \ , \tag{9}$$

where we have introduced the abbreviation

$$f(\mathbf{r};[\rho])\equiv\left.\frac{\delta T[\rho]}{\delta\rho(\mathbf{r})}\right|_{u,\mu} \ . \tag{10}$$

The solution of Eq. (9) gives the density $\rho(\mathbf{r})$ as a functional of $\bar{u}$, i.e., $\rho(\mathbf{r};[\bar{u}])$. It will be assumed that the quantity $\rho(\mathbf{r})$ thus obtained is a non-negative, integrable quantity for realistic potentials encountered in physical situations. The Lagrange multiplier μ is determined by the condition that the integrated density be equal to the prescribed number of particles, N:

$$\int d\mathbf{r}\rho(\mathbf{r};[\bar{u}])=N \ . \tag{11}$$

When $\rho(\mathbf{r})$ and μ as determined by Eqs. (9)–(11) are substituted into (7), the minimum value of Ω is obtained, and from it the ground-state energy E_N of the N-particle system via the relation

$$E_N=\Omega_{\min}+\mu N \ . \tag{12}$$

With the aid of Eqs. (12) and (8) it is easily verified that the Lagrange multiplier μ as determined via Eqs. (9)–(11) is in fact equal to the chemical potential defined by (6).

The above procedure is readily illustrated by the TFW approximation,[17] where the kinetic energy functional is approximated by the local functional

$$T_{\mathrm{TFW}}[\rho]=C_3\int d\mathbf{r}[\rho(\mathbf{r})]^{5/3}+\frac{\hbar^2}{8m}\int d\mathbf{r}\frac{[\nabla\rho(\mathbf{r})]^2}{\rho(\mathbf{r})} \tag{13}$$

(in three dimensions), where $C_3=3\hbar^2(3\pi^2)^{2/3}/(10m)$. In this case, Eqs. (8)–(10) lead to the nonlinear differential equation

$$\tfrac{5}{3}C_3[\rho(\mathbf{r})]^{2/3}+\frac{\hbar^2}{8m}\left[\left[\frac{\nabla\rho(\mathbf{r})}{\rho(\mathbf{r})}\right]^2-\frac{2\nabla_p^2(\mathbf{r})}{\rho(\mathbf{r})}\right]=-\bar{u}(\mathbf{r}) , \tag{14}$$

the solution of which determines $\rho_{\mathrm{TFW}}(\mathbf{r})$ as a function of $\bar{u}(\mathbf{r})$. This equation has been applied to a number of model problems, for example the Coulomb potential,[18] the harmonic oscillator, the Yukawa potential, and the Mathieu potential.[19]

As an aside, it is interesting to note that whereas relations (1)–(6) makes sense only when the particle number N is an integer, the analysis based on the density-functional approach, Eqs. (7)–(12), is defined even when N is no longer an integer. Thus, once the existence of the kinetic energy functional $T[\rho]$ is established, the variational procedure embodied by Eqs. (8)–(11) is well defined for any prescribed positive value of N. This fact is illustrated by the TFW approximation given above, where the solution $\rho_{\mathrm{TFW}}(\mathbf{r})$ of Eq. (14) and the determination of μ via (11) do not require that N be an integer.

From Eq. (9) we note that

$$\nabla u(\mathbf{r})=-\nabla f(\mathbf{r};[\rho]) . \tag{15}$$

Substituting (15) into the virial relation (5), we can write the kinetic energy functional $T[\rho]$ in the form given by Bartolotti and Parr,[12] Ghosh and Parr,[13] Levy and Perdew,[14] and Herring:[6]

$$T[\rho]=-\tfrac{1}{2}\int d\mathbf{r}\rho(\mathbf{r})\mathbf{r}\cdot\nabla f(\mathbf{r};[\rho]) . \tag{16}$$

Thus, if $f(\mathbf{r};[\rho])$ or any approximation to it is known, with $\rho(\mathbf{r})$ determined via Eq. (9), the corresponding $T[\rho]$ is determined. Equation (16) forms the basis of the hierarchy of equations to be derived in the next section.

III. EXACT HIERARCHY OF EQUATIONS

Functional differentiation with respect to $\rho(\mathbf{r})$ of both sides of Eq. (16) gives

$$f(\mathbf{r};[\rho])=-\tfrac{1}{2}\mathbf{r}\cdot\nabla f(\mathbf{r};[\rho]) -\tfrac{1}{2}\int d\mathbf{r}'\rho(\mathbf{r}')\mathbf{r}'\cdot\nabla' w_2(\mathbf{r},\mathbf{r}';[\rho]) , \tag{17}$$

where the two-point function $w_2(\mathbf{r},\mathbf{r}';[\rho])$ is defined as

$$w_2(\mathbf{r},\mathbf{r}';[\rho])=\frac{\delta f(\mathbf{r};[\rho])}{\delta\rho(\mathbf{r}')}=\frac{\delta^2 T[\rho]}{\delta\rho(\mathbf{r})\delta\rho(\mathbf{r}')} . \tag{18}$$

It is important to note that being the second functional derivative of $T[\rho]$, the quantity $w_2(\mathbf{r},\mathbf{r}'[\rho])$ is symmetric in its arguments $\mathbf{r}$ and $\mathbf{r}'$:

$$w_2(\mathbf{r},\mathbf{r}';[\rho])=w_2(\mathbf{r}',\mathbf{r};[\rho]) . \tag{19}$$

The density $\rho(\mathbf{r})$ which enters on both sides of Eq. (17) is determined by Eq. (9). Equation (17) constitutes a functional integrodifferential equation for $f(\mathbf{r};[\rho])$. We shall sometimes use the alternate form obtained from an integration by parts in the second term on the right-hand side of (17):

$$f(\mathbf{r};[\rho])=-\tfrac{1}{2}\mathbf{r}\cdot\nabla f(\mathbf{r};[\rho]) +\tfrac{1}{2}\int d\mathbf{r}'\nabla'\cdot[\mathbf{r}'\rho(\mathbf{r}')]w_2(\mathbf{r},\mathbf{r}';[\rho]) , \tag{20}$$

where it has been assumed that the product $\rho(\mathbf{r}')\mathbf{r}'w_2(\mathbf{r},\mathbf{r}';[\rho])$ vanishes at infinity, or at the bounding surface in the case of a system contained in a large, but finite volume V.

It is worth pointing out that the quantity $w_2(\mathbf{r},\mathbf{r}';[\rho])$ is closely related to the so-called "direct correlation" function $C(\mathbf{r},\mathbf{r}';[\rho],T)$ in a classical, interacting liquid, where $T[\rho]$ is replaced by the free energy functional $F[\rho,T]$ (T being the temperature).[20] In the present situation, $w_2(\mathbf{r},\mathbf{r}';[\rho])$ is in fact the matrix inverse of the static density-density response function, $\bar{\chi}_2(\mathbf{r},\mathbf{r}';[\bar{u}])$, for noninteracting fermions in the "potential" $\bar{u}(\mathbf{r})$; i.e., w_2 and $\bar{\chi}_2$ are related by

$$\int d\mathbf{r}' w_2(\mathbf{r},\mathbf{r}';[\rho])\bar{\chi}_2(\mathbf{r}',\mathbf{r}'';[\bar{u}])=\delta(\mathbf{r}-\mathbf{r}'') , \tag{21}$$

where it is understood that $\rho(\mathbf{r})$ and $\bar{u}(\mathbf{r})$ are related via Eqs. (9)–(11). This is shown explicitly in Sec. V. From Eqs. (8) and (18) we note the important fact that w_2 is equivalently given by

$$w_2(\mathbf{r},\mathbf{r}';[\rho])=\left.\frac{\delta^2\Omega}{\delta\rho(\mathbf{r})\delta\rho(\mathbf{r}')}\right|_{\bar{u}} . \tag{22}$$

Because the functional Ω is a minimum at the density $\rho(\mathbf{r})$ specified by Eq. (9), it follows that $w_2(\mathbf{r},\mathbf{r}';[\rho])$ must be a positive-definite matrix. This important property of w_2 is a consequence of, or requirement for, the stability of the system in the presence of the potential $u(\mathbf{r})$. This is further discussed in Sec. V.

Equations (16) and (17) constitute the zeroth and first of the equations in the hierarchy, respectively. Functional differentiation with respect to $\rho(\mathbf{r}')$ of Eq. (17) leads to the second equation:

$$w_2(\mathbf{r},\mathbf{r}';[\rho])=-\tfrac{1}{2}(\mathbf{r}\cdot\nabla+\mathbf{r}'\cdot\nabla')w_2(\mathbf{r},\mathbf{r}';[\rho])-\tfrac{1}{2}\int d\mathbf{r}''\rho(\mathbf{r}'')\mathbf{r}''\cdot\nabla''w_3(\mathbf{r},\mathbf{r}',\mathbf{r}'';[\rho]) \tag{23a}$$

$$=-\tfrac{1}{2}(\mathbf{r}\cdot\nabla+\mathbf{r}'\cdot\nabla')w_2(\mathbf{r},\mathbf{r}';[\rho])+\tfrac{1}{2}\int d\mathbf{r}''\nabla''\cdot[\mathbf{r}''\rho(\mathbf{r}'')]w_3(\mathbf{r},\mathbf{r}',\mathbf{r}'';[\rho]) , \tag{23b}$$

with (23b) following from integration by parts and where w_3 is the three-point function

$$w_3(\mathbf{r},\mathbf{r}',\mathbf{r}'';[\rho])=\frac{\delta w_2(\mathbf{r},\mathbf{r}';[\rho]}{\delta\rho(\mathbf{r}'')}=\frac{\delta^3 T[\rho]}{\delta\rho(\mathbf{r})\delta\rho(\mathbf{r}')\delta\rho(\mathbf{r}'')}\ . \tag{24}$$

It is now seen that further functional differentiation leads to successively higher-order equations. The equation relating the quantity

$$w_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;[\rho])\equiv\frac{\delta^n T[\rho]}{\delta\rho(\mathbf{r}_1)\cdots\delta\rho(\mathbf{r}_n)} \tag{25}$$

to $w_{n+1}(\mathbf{r}_1,\ldots,\mathbf{r}_{n+1};[\rho])$ reads

$$\begin{aligned}w_n(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_n;[\rho])=&-\tfrac{1}{2}(\mathbf{r}_1\cdot\nabla_1+\cdots+\mathbf{r}_n\cdot\nabla_n)w_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;[\rho])\\&-\tfrac{1}{2}\int d\mathbf{r}_{n+1}\rho(\mathbf{r}_{n+1})\mathbf{r}_{n+1}\cdot\nabla_{n+1}w_{n+1}(\mathbf{r}_1,\ldots,\mathbf{r}_{n+1};[\rho])\ .\end{aligned} \tag{26}$$

The quantity $w_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;[\rho])$ is symmetric with respect to interchange of any pair of its arguments, $\mathbf{r}_1\ldots\mathbf{r}_n$.

Equations (17), (23), (26), along with the "starting" relation (16) constitute the hierarchy of equations we set out to derive. It is of interest to note that these equations, valid for an inhomogeneous system of noninteracting fermions of density $\rho(\mathbf{r})$, bear a formal resemblance to the BBGKY hierarchy of equations in the theory of classical fluids which relate the interaction potential and successively higher-order distribution functions.[9,10]

In Sec. VI the validity of Eqs. (17) and (23) is demonstrated explicitly in the case of the uniform Fermi gas, where the quantities $f(\mathbf{r};[\rho])$, $w_2(\mathbf{r},\mathbf{r}';[\rho])$, and $w_3(\mathbf{r},\mathbf{r}',\mathbf{r}'';[\rho])$ can be obtained exactly.

IV. EXPRESSIONS FOR $T[\rho]$

With the aid of the hierarchy, the kinetic energy functional $T[\rho]$ can be expressed in several equivalent forms. First, using Eq. (17), the virial expression (16) can be rewritten in the forms

$$T[\rho]=\int d\mathbf{r}\,\rho(\mathbf{r})\{f(\mathbf{r};[\rho])+\tfrac{1}{2}\int d\mathbf{r}'\rho(\mathbf{r}')\mathbf{r}'\cdot\nabla' w_2(\mathbf{r},\mathbf{r}';[\rho])\} \tag{27a}$$

$$=\int d\mathbf{r}\,\rho(\mathbf{r})f(\mathbf{r};[\rho])+\tfrac{1}{4}\int\int d\mathbf{r}\,d\mathbf{r}'\rho(\mathbf{r})\rho(\mathbf{r}')(\mathbf{r}\cdot\nabla+\mathbf{r}'\cdot\nabla')w_2(\mathbf{r},\mathbf{r}';[\rho])\ , \tag{27b}$$

where, in going from (27a) to (27b), use was made of the property (19). Expressions (27) give the kinetic energy in terms of $\rho(\mathbf{r})$ and the one- and two-point functions, f and w_2, respectively. Equation (23a) can now be used to reexpress Eq. (27b) in terms of f, w_2, and w_3. Continuing in this way, one finally arrives at the following expression for $T[\rho]$ in the form of an infinite series, from which all the gradient terms have disappeared:

$$\begin{aligned}T[\rho]=&\int d\mathbf{r}\,\rho(\mathbf{r})f(\mathbf{r};[\rho])-\tfrac{1}{2}\int\int d\mathbf{r}\,d\mathbf{r}'\rho(\mathbf{r})\rho(\mathbf{r}')w_2(\mathbf{r},\mathbf{r}';[\rho])+\frac{1}{3!}\int d\mathbf{r}_1d\mathbf{r}_2d\mathbf{r}_3\rho(\mathbf{r}_1)\rho(\mathbf{r}_2)\rho(\mathbf{r}_3)w_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\rho])\\&-\frac{1}{4!}\int d\mathbf{r}_1d\mathbf{r}_2d\mathbf{r}_3d\mathbf{r}_4\rho(\mathbf{r}_1)\rho(\mathbf{r}_2)\rho(\mathbf{r}_3)\rho(\mathbf{r}_4)w_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4;[\rho])+\cdots\ .\end{aligned} \tag{28}$$

The alternating series (28) shows explicitly the contribution to the kinetic energy of the nth-order correlation function w_n and the densities associated with the points $\mathbf{r}_1,\ldots,\mathbf{r}_n$. (Note: Although it might at first be thought that the signs preceding the even terms $w_2,w_4,\ldots$ on the right-hand side of (28) should also be positive, this is not the case.) In Sec. VI we shall see that in the case of the uniform Fermi gas, the series (28) is rapidly converging, with the first three terms accounting for most of the kinetic energy of the uniform gas.

V. GENERAL RELATIONS INVOLVING THE QUANTITIES w_2 AND w_3

With the aid of Eqs. (8)–(10) it is easily verified that the functional defined by (7) yields the density $\rho(\mathbf{r};[\bar{u}])$ via functional differentiation with respect to $\bar{u}(\mathbf{r})$:

$$\left.\frac{\delta\Omega}{\delta\bar{u}(\mathbf{r})}\right|_{\rho}=\rho(\mathbf{r};[\bar{u}])\ . \tag{29}$$

Equations (7)–(10) and (29) indicate that $\rho(\mathbf{r})$ and $\bar{u}(\mathbf{r})$ are conjugate variables, and that from a formal standpoint these equations constitute a functional Legendre transformation from the set of quantities $\{T,\rho(\mathbf{r})\}$ to the set $\{\Omega,\bar{u}(\mathbf{r})\}$, and *vice versa*. We shall now use this fact to establish relations between the quantities $w_2(\mathbf{r},\mathbf{r}';[\rho])$, $w_3(\mathbf{r},\mathbf{r},\mathbf{r}'';[\rho]),\ldots$, and the so-called response functions $\chi_2(\mathbf{r},\mathbf{r}';[\bar{u}])$, $\chi_3(\mathbf{r},\mathbf{r}',\mathbf{r}'';[\bar{u}]),\ldots$ familiar from condensed matter physics. To establish the connection, we note that a change $\Delta u(\mathbf{r})$ in the potential also induces a change $\Delta\mu$ in the chemical potential. Thus we are led to consider the change in the functional $\Omega[\bar{u}]$ as $\bar{u}[\mathbf{r})$ is changed to $\bar{u}(\mathbf{r})+\Delta\bar{u}(\mathbf{r})$. By means of a functional Taylor expansion, we can write

$$\Omega[\bar{u}+\Delta\bar{u}]=\Omega[\bar{u}]+\int d\mathbf{r}\,\rho(\mathbf{r};[\bar{u}])\Delta\bar{u}(\mathbf{r})-\tfrac{1}{2}\int d\mathbf{r}_1 d\mathbf{r}_2\bar{\chi}_2(\mathbf{r}_1,\mathbf{r}_2;[\bar{u}])\Delta\bar{u}(\mathbf{r}_1)\Delta\bar{u}(\mathbf{r}_2)$$
$$-\frac{1}{3!}\int d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\bar{u}])\Delta\bar{u}(\mathbf{r}_1)\Delta\bar{u}(\mathbf{r}_2)\Delta\bar{u}(\mathbf{r}_3)-\cdots\ , \tag{30}$$

where [cf. Eq. (29)]

$$-\bar{\chi}_2(\mathbf{r}_1,\mathbf{r}_2;[\bar{u}])=\left.\frac{\delta^2\Omega[\bar{u}]}{\delta\bar{u}(\mathbf{r}_1)\delta\bar{u}(\mathbf{r}_2)}\right|_\rho=\frac{\delta\rho(\mathbf{r}_1;[\bar{u}])}{\delta\bar{u}(\mathbf{r}_2)}\ , \tag{31}$$

$$-\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\bar{u}])=\left.\frac{\delta^3\Omega[\bar{u}]}{\delta\bar{u}(\mathbf{r}_1)\delta\bar{u}(\mathbf{r}_2)\delta\bar{u}(\mathbf{r}_3)}\right|_\rho$$
$$=\frac{\delta^2\rho(\mathbf{r}_1;[\bar{u}])}{\delta\bar{u}(\mathbf{r}_2)\delta\bar{u}(\mathbf{r}_3)}\ . \tag{32}$$

The functions $\bar{\chi}_2$, $\bar{\chi}_3, \ldots$ are obviously symmetric under interchange of any pair of arguments. These functions are closely related to, but not identical with, the response functions obtainable from steady-state perturbation theory for a constant number of particles; expressions for the latter are given in Appendix A, where the exact relations between $\bar{\chi}_2(\mathbf{r}_1,\mathbf{r}_2;[\bar{u}])$, $\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\bar{u}])$, and the analogous functions for constant particle number N are obtained.

From the defining equation, (18), of $w_2(\mathbf{r},\mathbf{r}'[\rho])$, and from Eqs. (9) and (31), it follows immediately that

$$w_2(\mathbf{r},\mathbf{r}';[\rho])=\bar{\chi}_2^{-1}(\mathbf{r},\mathbf{r}';[\bar{u}])\ , \tag{33}$$

where ρ and $\bar{u}$ are related to each other via either Eq. (9) or via Eq. (29). The relation (33) is equivalent to (21).

The positive-definite property of $w_2(\mathbf{r},\mathbf{r}';[\rho])$, or equivalently of $\bar{\chi}_2(\mathbf{r},\mathbf{r}';[\bar{u}])$, will now be expressed in a more concrete way. First we note that the relation $\bar{u}(\mathbf{r})=u(\mathbf{r})-\mu$ implies the following relation between the partial derivative $\partial/\partial\mu|_u$ and the functional derivative $\delta/\delta\bar{u}(\mathbf{r})|_\rho$:

$$\left.\frac{\partial}{\partial\mu}\right|_u=-\int d\mathbf{r}\left.\frac{\delta}{\delta\bar{u}(\mathbf{r})}\right|_\rho\ . \tag{34}$$

Applying the operation (34) to the functional Ω, we have on account of (29)

$$\left.\frac{\partial\Omega}{\partial\mu}\right|_u=-\int d\mathbf{r}\,\rho(\mathbf{r};[\bar{u}])=-N\ , \tag{35}$$

a relation familiar from statistical mechanics. A second application of (34), this time to Eq. (35), yields

$$-\left.\frac{\partial^2\Omega}{\partial\mu^2}\right|_u=\int\int d\mathbf{r}\,d\mathbf{r}'\bar{\chi}_2(\mathbf{r},\mathbf{r}';[\bar{u}])=\left.\frac{\partial N}{\partial\mu}\right|_u\ . \tag{36}$$

Because of the positive-definite property of the matrix $\bar{\chi}_2$, we must have

$$\left.\frac{\partial N}{\partial\mu}\right|_u=\left.\frac{\partial\mu[u]}{\partial N}\right|_u^{-1}>0\ , \tag{37}$$

as a condition of stability of our system. A third application of (34), this time to the density $\rho(\mathbf{r})$, yields the more restrictive relation

$$\left.\frac{\partial\rho(\mathbf{r})}{\partial\mu}\right|_u=\int d\mathbf{r}'\bar{\chi}_2(\mathbf{r},\mathbf{r}';[\bar{u}])\ , \tag{38}$$

from which (36) is obtained via integration with respect to $\mathbf{r}$. This leads us to expect that

$$\left.\frac{\partial\rho(\mathbf{r})}{\partial\mu}\right|_u>0\ , \tag{39}$$

which is a more restrictive version of the inequality (37). Although not rigorously established at this point, with the aid of the relation

$$\left.\frac{\partial\rho(\mathbf{r})}{\partial\mu}\right|_u=\left.\frac{\partial\rho(\mathbf{r})}{\partial N}\frac{\partial N}{\partial\mu}\right|_u \tag{40}$$

(cf. also Appendix A), and the argument that $\rho(\mathbf{r})$ is a monotonically increasing function of the number of particles N [i.e., $\partial\rho(\mathbf{r})/\partial N>0$], inequality (39) follows from (37) and (40). Relations (38)–(40) are all rigorously verified for a uniform gas of density $\rho=N/V$, where they constitute the well-known "compressibility sum rule" familiar from linear response theory[21]

$$\int d\mathbf{r}'\bar{\chi}_2(\mathbf{r},\mathbf{r}';\mu)=\frac{\partial\rho}{\partial\mu}=\rho^2K>0\ , \tag{41}$$

K being the compressibility of the gas. For this system, an equivalent version of the compressibility sum rule can be given in terms of w_2, namely

$$\int d\mathbf{r}'w_2(\mathbf{r},\mathbf{r}';\rho)=\frac{\partial\mu}{\partial\rho}=\frac{1}{\rho^2K}\ . \tag{42}$$

Equation (42) follows from Eqs. (33) and (41) and the spatial invariance of the uniform system. Relations (41) and (42) will be used in the next section.

Via the chain rule for functional differentiation applied to Eq. (21), we obtain the equation connecting the quantity $w_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\rho])$ with the quantity $\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\bar{u}])$:

$$w_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\rho])=\int d\mathbf{r}_4 d\mathbf{r}_5 d\mathbf{r}_6\bar{\chi}_3(\mathbf{r}_4,\mathbf{r}_5,\mathbf{r}_6;[\bar{u}])\bar{\chi}_2^{-1}(\mathbf{r}_4,\mathbf{r}_1;[\bar{u}])\bar{\chi}_2^{-1}(\mathbf{r}_5,\mathbf{r}_2;[\bar{u}])\bar{\chi}_2^{-1}(\mathbf{r}_6,\mathbf{r}_3;[u])$$
$$=\int d\mathbf{r}_4 d\mathbf{r}_5 d\mathbf{r}_6\bar{\chi}_3(\mathbf{r}_4,\mathbf{r}_5,\mathbf{r}_6;[\bar{u}])w_2(\mathbf{r}_4,\mathbf{r}_1;[\rho])w_2(\mathbf{r}_5,\mathbf{r}_2;[\rho])w_2(\mathbf{r}_6,\mathbf{r}_3;[\rho])\ . \tag{43}$$

This relation will be needed in the next section. The chain rule can now be applied to Eq. (43) and the process continued to obtain the equations relating the higher-order correlation functions $w_4, w_5 \ldots$ to the quantities $\bar{\chi}_4, \bar{\chi}_5 \cdot \ldots$

VI. EQUATIONS FOR THE UNIFORM (HOMOGENEOUS) GAS

We now demonstrate explicitly the validity of the first and second equations of the hierarchy, Eqs. (17) and (23), in the uniform gas case. Relations and expressions will be given for both the three-dimensional ($d=3$) gas and the one-dimensional ($d=1$) gas; the latter case is also of interest in connection with Herring's work.[6]

In the case of the noninteracting, uniform gas, $u(\mathbf{r})=0, \bar{u}(\mathbf{r})=-\mu$, the density $\rho(\mathbf{r})$ has the uniform value $\rho=N/V$, where V is the volume of the container. In this case, because of spatial invariance, the quantity $f(\mathbf{r};[\rho])$ defined by (10) has the uniform value (except very close to the surface)

$$f(\mathbf{r};[\rho])=f_0(\rho)=\frac{d}{d\rho}t_0(\rho) , \tag{44}$$

where

$$t_0(\rho)=\begin{cases} C_3\rho^{5/3}, \quad C_3=\dfrac{3\hbar^2}{10m}(3\pi^2)^{2/3}, \quad d=3 , & (45)\\[2ex] C_1\rho^3, \quad C_1=\dfrac{\hbar^2\pi^2}{24m}, \quad d=1 . & (46)\end{cases}$$

Hence

$$f_0(\rho)=\begin{cases} \frac{5}{3}C_3\rho^{2/3}=\dfrac{\hbar^2k_F^2}{2m}, \quad k_F^3=3\pi^2\rho, d=3 , & (47)\\[2ex] 3C_1\rho^2=\dfrac{\hbar^2k_F^2}{2m}, \quad k_F=\dfrac{\pi\rho}{2}, d=1 . & (48)\end{cases}$$

The quantity $\hbar^2k_F^2/(2m)$ is equal to the chemical potential μ in both three-dimensional and one-dimensional gas, respectively, and hence Eqs. (47) and (48) express the content of Eq. (9) for the uniform gas, i.e.,

$$f_0(\rho)=\mu . \tag{49}$$

To show the validity of Eq. (17), we start from the equivalent version (20). The first term on the right-hand side of (20) does not contribute here, while the second term gives

$$\tfrac{1}{2}\int d\mathbf{r}'\nabla'\cdot[\mathbf{r}'\rho(\mathbf{r}')]w_2(\mathbf{r},\mathbf{r}';[\rho])=\frac{\rho d}{2}\int d\mathbf{r}'w_2(\mathbf{r},\mathbf{r}';\rho) , \tag{50}$$

where

$$w_2(\mathbf{r},\mathbf{r}';\rho)=\left.\frac{\delta^2T[\rho]}{\delta\rho(\mathbf{r})\delta\rho(\mathbf{r}')}\right|_{\rho(\mathbf{r})=\rho} . \tag{51}$$

On account of spatial invariance, w_2 is a function only of the difference $\mathbf{r}-\mathbf{r}'$; thus, we can use the Fourier transform

$$\begin{aligned} w_2(\mathbf{r}-\mathbf{r}';\rho)&=\frac{1}{V}\sum_{\mathbf{k}} w(k;\rho)e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\\ &\to \int d\mathbf{k}(2\pi)^{-d}w(k;\rho)e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} , \quad k=|\mathbf{k}| . \end{aligned} \tag{52}$$

Furthermore, as follows in this case from (33),

$$w(k;\rho)=\frac{1}{\bar{\chi}(k;\rho)} , \tag{53}$$

where $\bar{\chi}(k;\rho)$ is the well-known linear response function of the uniform gas given by the Lindhard[15] expressions

$$\bar{\chi}(k,\rho)=\begin{cases} \dfrac{mk_F}{2\pi^2\hbar^2}\left[1+\dfrac{1-\eta^2}{2\eta}\ln\left|\dfrac{1+\eta}{1-\eta}\right|\right], \quad d=3 , & (54)\\[2ex] \dfrac{m}{\pi\hbar^2k_F}\dfrac{1}{\eta}\ln\left|\dfrac{1+\eta}{1-\eta}\right|, \quad d=1 , & (55)\end{cases}$$

where $\eta=k/(2k_F)$. The function $w_2(\mathbf{r}-\mathbf{r}';\rho)$ defined by Eqs. (52)–(55) is essentially the same as that considered by Herring.[6] (This is discussed further at the end of this section). Using the Fourier-transform relations (52) and (53), the right-hand side of (50) becomes

$$\begin{aligned} \frac{\rho d}{2}\int d\mathbf{r}'w_2(\mathbf{r}-\mathbf{r}';\rho)&=\frac{\rho d}{2}\lim_{k\to0}w(k;\rho)\\ &=\frac{\rho d}{2}\lim_{k\to0}\frac{1}{\bar{\chi}(k;\rho)} . \end{aligned} \tag{56}$$

From expressions (54) and (55) one obtains

$$\lim_{k\to0}\bar{\chi}(k;\rho)\equiv\bar{\chi}(k\to0;\rho)=\begin{cases} \dfrac{mk_F}{\pi^2\hbar^2}, \quad d=3 & (57)\\[2ex] \dfrac{2m}{\pi\hbar^2k_F}, \quad d=1 . & (58)\end{cases}$$

When it is recalled that $\mu=\hbar^2k_F^2/(2m)$, from which $\partial\mu/\partial\rho=2\mu/(\rho d)$, Eqs. (57) and (58) are seen to reduce to the single statement, valid for both $d=3$ and $d=1$,

$$\bar{\chi}(k\to0;\rho)=\frac{\partial\rho}{\partial\mu} , \tag{59}$$

which is consistent with Eq. (41). Substituting the values (57) and (58) into Eq. (56), and taking into account the relations between k_F and ρ [Eqs. (47) and (48)] yields

$$\frac{\rho d}{2}\int d\mathbf{r}'w_2(\mathbf{r}-\mathbf{r}';\rho)=\frac{\hbar^2k_F^2}{2m}=f_0(\rho) , \tag{60}$$

for $d=3$ and $d=1$, respectively, thus verifying explicitly the first equation of the hierarchy. Equation (60) is thus only a disguised version of Eq. (41) or Eq. (42).

Next we wish to verify the second equation of the hierarchy, Eq. (23). For the spatially invariant, uniform system, Eq. (23b) becomes

$$w_2(\mathbf{r}-\mathbf{r}';\rho)=-\tfrac{1}{2}(\mathbf{r}-\mathbf{r}')\cdot\nabla w_2(\mathbf{r}-\mathbf{r}';\rho)$$
$$+\frac{\rho d}{2}\int d\mathbf{r}''w_3(\mathbf{r},\mathbf{r}',\mathbf{r}'';\rho)\ , \tag{61}$$

where

$$w_3(\mathbf{r},\mathbf{r}',\mathbf{r}'',\rho)=\left.\frac{\delta^3T[\rho]}{\delta\rho(\mathbf{r})\delta\rho(\mathbf{r}')\delta\rho(\mathbf{r}'')}\right|_{\rho(\mathbf{r}_l)=\rho}\ , \tag{62}$$

is a function only of the coordinate differences, $\mathbf{r}-\mathbf{r}''$ and $\mathbf{r}'-\mathbf{r}''$.

To establish the equality of both sides of (61) explicitly, it is convenient to put $\mathbf{r}'\equiv 0$ and to work with the Fourier component [cf. Eq. (52)]

$$w(k;\rho)=\int d\mathbf{r}\,w_2(\mathbf{r};\rho)e^{-i\mathbf{k}\cdot\mathbf{r}}\ . \tag{63}$$

Equation (61) is then equivalent to the equation

$$w(k;\rho)=\frac{d}{2}w(k;\rho)+\frac{\mathbf{k}}{2}\cdot\nabla_{\mathbf{k}}w(k;\rho)$$
$$+\frac{\rho d}{2}\int\int d\mathbf{r}\,d\mathbf{r}'e^{-i\mathbf{k}\cdot\mathbf{r}}w_3(\mathbf{r},\mathbf{r}',0;\rho)\ . \tag{64a}$$

Collecting all the terms involving $w(k;\rho)$ on the left-hand side, the equation to be proved becomes

$$w(k;\rho)\left[2-d-k\frac{\partial}{\partial k}\ln w(k;\rho)\right]$$
$$=\rho d\int\int d\mathbf{r}\,d\mathbf{r}'e^{-i\mathbf{k}\cdot\mathbf{r}}w_3(\mathbf{r},\mathbf{r}',0;\rho)\ . \tag{64b}$$

Evaluation of the right-hand side of (64b) requires use of the identity (43) applied to the uniform gas; for the latter, Eq. (43) becomes

$$w_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;\rho)=\int d\mathbf{r}_4d\mathbf{r}_5d\mathbf{r}_6\bar{\chi}_3(\mathbf{r}_4,\mathbf{r}_5,\mathbf{r}_6;\rho)w_2(\mathbf{r}_4,\mathbf{r}_1;\rho)w_2(\mathbf{r}_5,\mathbf{r}_2;\rho)w_2(\mathbf{r}_6,\mathbf{r}_3;\rho)\ . \tag{65}$$

The quantity $\bar{\chi}_3(4,5,6;\rho)$ is the quadratic density response function of the uniform gas. Using identity (65) we have

$$\int d\mathbf{r}_1d\mathbf{r}_2e^{-i\mathbf{k}\cdot\mathbf{r}_1}w_3(\mathbf{r}_1,\mathbf{r}_2,0;\rho)=w(k\to 0;\rho)w(k;\rho)\int d\mathbf{r}_4d\mathbf{r}_5,d\mathbf{r}_6\bar{\chi}_3(\mathbf{r}_4,\mathbf{r}_5,\mathbf{r}_6;\rho)e^{-i\mathbf{k}\cdot\mathbf{r}_4}w(\mathbf{r}_6,0;\rho)$$
$$=w(k\to 0;\rho)w(k;\rho)\frac{1}{V}\sum_{k'}\bar{\chi}_3(\mathbf{k},\mathbf{k}'0;\rho)w(-\mathbf{k}';\rho)\ , \tag{66}$$

where $\bar{\chi}_3(\mathbf{k},\mathbf{k}',\mathbf{k}'';\rho)$ is the Fourier transform defined by

$$\bar{\chi}_3(\mathbf{k},\mathbf{k}',\mathbf{k}'';\rho)=\int d\mathbf{r}_1d\mathbf{r}_2d\mathbf{r}_3\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;\rho)e^{-i(\mathbf{k}\cdot\mathbf{r}_1+\mathbf{k}'\cdot\mathbf{r}_2+\mathbf{k}''\cdot\mathbf{r}_3)}\ . \tag{67}$$

The quantity $\bar{\chi}_3(\mathbf{k},\mathbf{k}',\mathbf{k}'';\rho)$ is symmetric with respect to interchange of any pair of its arguments and, because of spatial invariance, vanishes unless $\mathbf{k}+\mathbf{k}'+\mathbf{k}''=0$. Therefore, only the term $\mathbf{k}'=-\mathbf{k}$ contributes in the summation on the right-hand side of Eq. (66) and the latter becomes

$$\int d\mathbf{r}_1d\mathbf{r}_2e^{-i\mathbf{k}\cdot\mathbf{r}_1}w_3(\mathbf{r}_1,\mathbf{r}_2,0;\rho)=w(k\to 0;\rho)[w(k;\rho)]^2\frac{1}{V}\bar{\chi}_3(\mathbf{k},-\mathbf{k},0;\rho)\ . \tag{68}$$

Hence Eq. (64b) is equivalent to the relation

$$\left[2-d-k\frac{\partial}{\partial k}\ln w(k;\rho)\right]=\rho d w(k\to 0;\rho)w(k;\rho)\frac{1}{V}\bar{\chi}_3(\mathbf{k},-\mathbf{k},0;\rho)\ , \tag{69}$$

or, with (53)

$$\bar{\chi}(k\to 0;\rho)\bar{\chi}(k;\rho)\left[2-d+\frac{k\bar{\chi}'(k;\rho)}{\bar{\chi}(k;\rho)}\right]=\frac{\rho d}{V}\bar{\chi}_3(\mathbf{k},-\mathbf{k},0;\rho)\ , \tag{70}$$

where $k\bar{\chi}'(k;\rho)\equiv k\partial\bar{\chi}(k;\rho)/\partial k=\eta\partial\bar{\chi}(k;\rho)/\partial\eta$. The left-hand side of (70) is easily evaluated using expressions (54) and (55), and one finds

$$\bar{\chi}(k\to 0;\rho)\bar{\chi}(k;\rho)\left[2-d+\frac{k\bar{\chi}'(k;\rho)}{\bar{\chi}(k;\rho)}\right]=\begin{cases}-\left[\dfrac{mk_F}{\pi^2\hbar^2}\right]\dfrac{1}{2\eta}\ln\left|\dfrac{1+\eta}{1-\eta}\right|, & d=3\ , \quad (71)\\[2ex] \left[\dfrac{2m}{\pi\hbar^2k_F}\right]^2\dfrac{1}{1-\eta^2}, & d=1\ . \quad (72)\end{cases}$$

Evaluation of the right-hand side of (70) first involves establishing the identity

$$\frac{\rho d}{V}\bar{\chi}_3(\mathbf{k},-\mathbf{k},0;\rho)=-\rho d\bar{\chi}(k\to 0;\rho)\frac{\partial}{\partial\rho}\bar{\chi}(k;\rho) \tag{73a}$$
$$=-\bar{\chi}(k\to 0;\rho)k_F\frac{\partial}{\partial k_F}\bar{\chi}(k;\rho)\ , \tag{73b}$$

where relations (47) and (48) between ρ and k_F have been used in going from (73a) to (73b). The proof of identity (73a) is nontrivial and is given in detail in Appendix B. The right-hand side of Eq. (73b) is now easily evaluated using expressions (54) and (55) and is found to be identical with (71) and (72) for $d=3$ and $d=1$, respectively. The validity of Eqs. (61) and (64) is therefore established.

For the uniform gas, the general equation (26) linking w_n to w_{n+1} becomes

$$w_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;\rho)=-\tfrac{1}{2}(\mathbf{r}_1\cdot\nabla_1+\cdots+\mathbf{r}_n\cdot\nabla_n)w_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;\rho)+\frac{\rho d}{2}\int d\mathbf{r}_{n+1}w_{n+1}(\mathbf{r}_1,\ldots,\mathbf{r}_n,\mathbf{r}_{n+1};\rho)\ . \tag{74}$$

Equation (74) will prove useful below.

We next consider the expressions for the kinetic energy functional derived in Sec. IV. For the uniform gas, we obtain from the general infinite series expression (28), after substituting the quantities $f_0(\rho)$, $w_2(\mathbf{r}-\mathbf{r}';\rho)$, $w_3(1,2,3;\rho)\ldots$, the following expression for the kinetic energy $T_0(\rho)$:

$$\begin{aligned}T_0(\rho)=&\int d\mathbf{r}\rho f_0(\rho)-\frac{\rho^2}{2}\int d\mathbf{r}_1 d\mathbf{r}_2 w_2(\mathbf{r}_1,\mathbf{r}_2;\rho)+\frac{\rho^3}{3!}\int d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3 w_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;\rho)\\&-\frac{\rho^4}{4!}\int d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3 d\mathbf{r}_4 w_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4;\rho)+\cdots\ ,\end{aligned} \tag{75}$$

or, making use of spatial invariance, and dividing both sides by the volume,

$$t_0(\rho)=\rho f_0(\rho)-\frac{\rho^2}{2}\int d\mathbf{r}_2 w_2(\mathbf{r}_1,\mathbf{r}_2;\rho)+\frac{\rho^3}{3!}\int d\mathbf{r}_2 d\mathbf{r}_3 w_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;\rho)-\cdots\ , \tag{76}$$

where $t_0(\rho)$ is given by (45) and (46) for $d=3$ and $d=1$, respectively. Equation (76) may be regarded as a sum rule for the integrated quantities $w_2, w_3\ldots$ which represent the contributions to the kinetic energy from two-point, three-point, and higher-order correlations.

We can use the hierarchy equation (74) to evaluate the contribution to $t_0(\rho)$ from the $(n+1)$th term in (76) in terms of the nth term. One easily finds

$$\frac{\rho^{n+1}}{(n+1)!}\int d\mathbf{r}_2\cdots d\mathbf{r}_{n+1}w_{n+1}(\mathbf{r}_1,\ldots,\mathbf{r}_{n+1};\rho)=\frac{1}{(n+1)}\left[\frac{2}{d}-(n-1)\right]\frac{\rho^n}{n!}\int d\mathbf{r}_2\cdots d\mathbf{r}_n w_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;\rho)\ . \tag{77}$$

Hence, starting with $n=1$ we can obtain successively the contributions of each term in the series expression (76). In this way we find that the first five terms of the series (76) give, for $d=3$,

$$\tfrac{5}{3}C_3\rho^{5/3}\left(1-\tfrac{1}{3}-\tfrac{1}{27}-\tfrac{1}{81}-\tfrac{7}{1215}-\cdots\right)=C_3\rho^{5/3}\tfrac{1253}{1215}\ . \tag{78}$$

Comparing (78) with (45), we see that the coefficient $\frac{1253}{1215}$ is only 3% larger than the exact value, 1; in fact, even the first three terms alone yield a coefficient, $\frac{85}{81}$, which is only 5% larger than the exact value. This suggests that the series (76) converges rapidly for the-three dimensional gas.

In the one-dimensional gas, we observe from (77) that the coefficient on the right vanishes when $n=3$, leading to

$$\int d\mathbf{r}_2 d\mathbf{r}_3 d\mathbf{r}_4 w_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4)=0,\quad d=1\ . \tag{79}$$

Thus, all the higher-order contributions, starting with w_4, do not contribute to the series (76) and we are left with the contribution from the first three terms, which give

$$3C_1\rho^3\left(1-1+\tfrac{1}{3}\right)=C_1\rho^3,\quad d=1\ , \tag{80}$$

i.e., we obtain the exact value for $t_0(\rho)$ [compare (46)], as expected. From (77) it is also noted that in the two-dimensional gas, a similar situation occurs when $n=2$.

The termination of the series (76) for $d=1$ and $d=2$ appears to result from special features connected with these systems and suggests that the structure of the functional $T[\rho]$ is much simpler for the one- and two-dimensional gas than for the three-dimensional gas. This will not be explored further in this work.

Finally, we give the relation of the function $w_2(\mathbf{r}-\mathbf{r}';\rho)$ to the function $Q(2k_F|\mathbf{r}-\mathbf{r}'|)$ introduced and evaluated by Herring.[6] The quantity $w(k;\rho)$ defined by Eqs. (52)–(55) behaves asymptotically for large k, i.e., $\eta\gg 1$, as

$$w(k;\rho)=\begin{cases}\dfrac{2\pi^2\hbar^2}{mk_F}\left[\dfrac{3\eta^2}{2}-\dfrac{3}{10}-\dfrac{12}{175\eta^2}\cdots\right], & d=3\ , \qquad (81)\\[2ex] \dfrac{\pi\hbar^2 k_F}{m}\left[\dfrac{\eta^2}{2}-\dfrac{1}{6}-\dfrac{2}{45\eta^2}\cdots\right], & d=1\ . \qquad (82)\end{cases}$$

As was already pointed out by Jones and Young,[17] the leading term is that which is obtained from the von Weizsäcker approximation for the energy of a weakly perturbed Fermi gas. The asymptotic expressions (81) and (82) allow us to write $w_2(\mathbf{r}-\mathbf{r}';\rho)$ as a sum of local and nonlocal contributions, i.e.,

$$\begin{aligned}w_2(\mathbf{r}-\mathbf{r}';\rho)=&\,q_0(\rho)\delta(\mathbf{r}-\mathbf{r}')-q_2(\rho)\nabla^2\delta(\mathbf{r}-\mathbf{r}')\\&+w_{\mathrm{nl}}(\mathbf{r}-\mathbf{r}';\rho)\ ,\end{aligned} \tag{83}$$

where

$$q_0(\rho)=\begin{cases} -\dfrac{3\pi^2\hbar^2}{5mk_F}=-\dfrac{\hbar^2k_F^2}{5m\rho}, \quad d=3\ , & (84)\\[2ex] -\dfrac{\pi\hbar^2k_F}{6m}=-\dfrac{\hbar^2k_F^2}{3m\rho}, \quad d=1\ , & (85)\end{cases}$$

$$q_2(\rho)=\begin{cases} \dfrac{3\pi^2\hbar^2}{4mk_F^3}=\dfrac{\hbar^2}{4m\rho}, \quad d=3 & (86)\\[2ex] \dfrac{\pi\hbar^2}{8mk_F}=\dfrac{\hbar^2}{4m\rho}, \quad d=1\ . & (87)\end{cases}$$

and the nonlocal part, $w_{\mathrm{nl}}(\mathbf{r}-\mathbf{r}';\rho)$ is given by

$$w_{\mathrm{nl}}(\mathbf{r}-\mathbf{r}';\rho)=\begin{cases} \dfrac{2\pi^2\hbar^2}{mk_F}\left[\dfrac{k_F}{\pi}\right]^3\displaystyle\int d^3\eta\, e^{i\boldsymbol{\eta}\cdot\mathbf{y}}\left[\left[1+\frac{(1-\eta^2)}{2\eta}\ln\left|\frac{1+\eta}{1-\eta}\right|\right]^{-1}-\frac{3\eta^2}{2}+\frac{3}{10}\right], \quad d=3\ , & (88)\\[3ex] \dfrac{\pi\hbar^2k_F}{m}\left[\dfrac{k_F}{\pi}\right]\displaystyle\int d\eta\, e^{i\eta y}\left[\frac{\eta}{\ln\left|\dfrac{1+\eta}{1-\eta}\right|}-\frac{\eta^2}{2}+\frac{1}{6}\right], \quad d=1\ , & (89)\end{cases}$$

where $\mathbf{y}\equiv 2k_F(\mathbf{r}-\mathbf{r}')$. Except for a numerical factor, the functions w_{nl} are identical with the functions denoted $Q(u)$ by Herring.

VII. CONCLUDING REMARKS

Starting from the virial theorem and the fact that the kinetic energy is a unique functional of the particle density, we have derived a hierarchy of coupled, exact equations which in principle completely determine the structure of this functional for an arbitrary, inhomogeneous system of noninteracting fermions. The equations themselves do not involve the external potential, nor the number of particles in the system, but only the particle density and quantities which are closely related to the functions which describe the static response of the system to a change in the potential.

The challenge and task now remain to apply these equations to obtain more accurate functional expressions for $T[\rho]$ and its first few functional derivatives. Not surprisingly, the degree of complexity involved in using the hierarchy is analogous to that encountered in the BBGKY hierarchy for nonuniform, classical fluids, a topic of ongoing investigations.[22]

It is hoped that the equations presented here will provide a rigorous basis for, and prove useful in, comparing and testing various forms of local and nonlocal functionals of the density and in deriving new expressions for the latter. On the other hand, since the equations themselves contain no reference to local, versus nonlocal, terms one might also bypass any separation into such terms and attempt to focus on getting expressions for the quantities as they have been defined here. In any event, it will be necessary to introduce approximations, such as truncating the hierarchy at a given order and then attempting a solution of the closed system of equations. If such an approach is attempted, one may be guided to use only approximation schemes which are exact in the uniform gas case discussed in Sec. VI (see also Appendix B). In pursuing such work, it will undoubtedly also prove useful to draw on the experience gained and lessons learned in dealing with classical, nonuniform systems.

In addition to the above, it will be of interest to investigate the extent to which the ideas presented here can be used for the case of real, interacting fermions, in particular the general problem of electrons in atoms, molecules, and solids. It is hoped that studies will be carried out along these lines.

APPENDIX A: RELATIONS BETWEEN THE QUANTITIES $\bar{\chi}(\mathbf{r}_1,\mathbf{r}_2,\ldots,[\bar{u}])$ AND THE N-PARTICLE RESPONSE FUNCTIONS $\chi(\mathbf{r}_1,\mathbf{r}_2,\ldots;[u])$ OF AN INHOMOGENEOUS FERMI GAS

Here we wish to relate the quantities $\bar{\chi}_2$ and $\bar{\chi}_3$ introduced in Sec. V to the corresponding quantities which describe the linear and quadratic response of an N-particle system to a change $\Delta u(\mathbf{r})$ in the potential. That is, assuming the system is described initially by the Hamiltonian (1), we apply the perturbation

$$\Delta\hat{U}=\sum_{i=1}^{N}\Delta u(\mathbf{r}_i)=\int d\mathbf{r}\hat{\rho}(\mathbf{r})\Delta u(\mathbf{r})\ , \tag{A1}$$

and ask for the change in the ground-state energy resulting from this perturbation. It is important to note here that no condition is placed on the perturbation $\Delta\hat{U}$ other than that it maintain the stability of the initial system. In particular, we do not impose the condition that the integral of $\Delta u(\mathbf{r})$ over the volume occupied by the system (in the event that the latter is finite) vanish. Such a condition is usually imposed in the case of a nearly uniform gas treated within the linear response regime; this then has the consequence that to first order in Δu, the chemical potential remains unchanged from its initial, unperturbed value. No such condition is made here.

We shall first write the energy corresponding to the potential $u(\mathbf{r})+\Delta u(\mathbf{r})$ formally as a functional Taylor series, analogous to (30):

$$E_N(u+\Delta u)=E_N[u]+\int d\mathbf{r}\,\rho(\mathbf{r})\Delta u(\mathbf{r})-\tfrac{1}{2}\int d\mathbf{r}_1 d\mathbf{r}_2\,\chi_2(\mathbf{r}_1,\mathbf{r}_2;[u])\Delta u(\mathbf{r}_1)\Delta u(\mathbf{r}_2)$$
$$-\frac{1}{3!}\int d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3\,\chi_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[u])\Delta u(\mathbf{r}_1)\Delta u(\mathbf{r}_2)\Delta u(\mathbf{r}_3)-\cdots\;, \tag{A2}$$

where

$$\rho(\mathbf{r})=\rho(\mathbf{r};[u])=\left.\frac{\delta E_N[u]}{\delta u(\mathbf{r})}\right|_N\;, \tag{A3}$$

$$-\chi_2(\mathbf{r}_1,\mathbf{r}_2;[u])=\left.\frac{\delta^2 E_N[u]}{\delta u(\mathbf{r}_1)\delta u(\mathbf{r}_2)}\right|_N=\left.\frac{\delta\rho(\mathbf{r}_1)}{\delta u(\mathbf{r}_2)}\right|_N\;, \tag{A4}$$

$$-\chi_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[u])=\left.\frac{\delta^3 E_N[u]}{\delta u(\mathbf{r}_1)\delta u(\mathbf{r}_2)\delta u(\mathbf{r}_3)}\right|_N=\left.\frac{\delta^2\rho(\mathbf{r}_1)}{\delta u(\mathbf{r}_2)\delta u(\mathbf{r}_3)}\right|_N\;. \tag{A5}$$

It is understood here that all functional derivatives above are taken at constant particle number N. The quantities $\chi(\mathbf{r}_1,\mathbf{r}_2,\ldots;[u])$ are symmetric with respect to interchange of any pair of arguments. χ_2 and χ_3 are usually referred to as the linear and quadratic response functions, respectively, since the change Δu leads to a change $\Delta\rho$ in the density, given to second order in Δu by

$$\Delta\rho(\mathbf{r}_1)=-\int d\mathbf{r}_2\,\chi_2(\mathbf{r}_1,\mathbf{r}_2;[u])\Delta u(\mathbf{r}_2)-\tfrac{1}{2}\int d\mathbf{r}_2 d\mathbf{r}_3\,\chi_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[u])\Delta u(\mathbf{r}_2)\Delta u(\mathbf{r}_3)\;. \tag{A6}$$

Assuming that the eigenstates of the system with the potential $u+\Delta u$ are only slightly different from the original eigenstates corresponding to u, we can apply the usual stationary state perturbation theory[23] to obtain $E_N[u+\Delta u]$. To third order in $\Delta\hat{U}$, the perturbation expansion gives

$$E_N(u+\Delta u)=E_N[u]+\langle 0|\Delta\hat{U}|0\rangle-\sum_n{}'\frac{\langle 0|\Delta\hat{U}|n\rangle\langle n|\Delta\hat{U}|0\rangle}{E_n-E_0}+\sum_n{}'\sum_m{}'\frac{\langle 0|\Delta\hat{U}|n\rangle\langle n|\Delta\hat{U}|m\rangle\langle m|\Delta\hat{U}|0\rangle}{(E_n-E_0)(E_m-E_0)}$$
$$-\langle 0|\Delta\hat{U}|0\rangle\sum_n{}'\frac{\langle 0|\Delta\hat{U}|n\rangle\langle n|\Delta\hat{U}|0\rangle}{(E_n-E_0)^2}\;, \tag{A7}$$

where the quantities $E_0=E_N[u]$, $E_m,E_n\ldots$ denote, respectively, the energy of the ground state $|0\rangle\equiv|\Phi_0[u]\rangle$ and excited states $|m\rangle,|n\rangle,\ldots,$ and the primes on the summation symbols imply that the summations are to exclude the ground state $|0\rangle$. The states $|0\rangle,|m\rangle,\ldots,$ are N-particle determinants of one-particle eigenstates (cf. below). When (A1) is substituted into (A7), comparison with (A2) yields

$$\rho(\mathbf{r})=\langle 0|\hat{\rho}(\mathbf{r})|0\rangle\;, \tag{A8}$$

$$\tfrac{1}{2}\chi_2(\mathbf{r}_1,\mathbf{r}_2;[u])=\sum_n{}'\frac{\langle 0|\hat{\rho}(\mathbf{r}_1)|n\rangle\langle n|\hat{\rho}(\mathbf{r}_2)|0\rangle}{E_n-E_0}\;, \tag{A9}$$

$$-\frac{1}{3!}\chi_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[u])=\sum_n{}'\sum_m{}'\frac{\langle 0|\hat{\rho}(\mathbf{r}_1)|n\rangle\langle n|\hat{\rho}(\mathbf{r}_3)|m\rangle\langle m|\hat{\rho}(\mathbf{r}_2)|0\rangle}{(E_n-E_0)(E_m-E_0)}$$
$$-\sum_n{}'\frac{\langle 0|\hat{\rho}(\mathbf{r}_1)|n\rangle\langle n|\hat{\rho}(\mathbf{r}_2)|0\rangle\langle 0|\hat{\rho}(\mathbf{r}_3)|0\rangle}{(E_n-E_0)^2}\;. \tag{A10}$$

A useful property which follows from (A9) and (A10) is that the integrals of χ_2 and χ_3 with respect to any of their arguments vanish; thus

$$\int d\mathbf{r}_1\,\chi_2(\mathbf{r}_1,\mathbf{r}_2;[u])=0\;, \tag{A11}$$

$$\int d\mathbf{r}_1\,\chi_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[u])=0\;. \tag{A12}$$

[This is easily seen by noting that the integral of $\hat{\rho}(\mathbf{r})$ gives the c number N and then invoking the orthonormality of the states occurring in the sums.] Equations (A11) and (A12) also follow from the normalization condition

$$\int d\mathbf{r}\,\rho(\mathbf{r}_i[u])=N\;, \tag{A13}$$

by repeated application of the functional derivative with respect to u at constant N [cf. (A4) and (A5)].

It is instructive to express $\rho(\mathbf{r})$ and $\chi_2(\mathbf{r}_1,\mathbf{r}_2);\,[u])$ explicitly in terms of the single-particle eigenfunctions $\varphi_i(\mathbf{r})$ satisfying the Schrödinger equation

$$\left[-\frac{\hbar^2}{2m}\nabla^2+u(\mathbf{r})\right]\varphi_i(\mathbf{r})=\varepsilon_i\varphi_i(\mathbf{r}) , \tag{A14}$$

where ε_i is the associated energy level. In terms of these quantities, (A8) and (A9) become

$$\rho(\mathbf{r})=\sum_{i,s} n_s(\varepsilon_i)|\varphi_i(\mathbf{r})|^2 , \tag{A15}$$

$$\begin{aligned}\chi_2(\mathbf{r}_1,\mathbf{r}_2;[u])&=2\sum_{\substack{i,j,s\\ i\neq j}}\frac{n_s(\varepsilon_j)[1-n_s(\varepsilon_i)]}{\varepsilon_i-\varepsilon_j}\varphi_i(\mathbf{r}_1)\varphi_i^*(\mathbf{r}_2)\varphi_j^*(\mathbf{r}_1)\varphi_j(\mathbf{r}_2)\\ &=\sum_{\substack{i,j,s\\ i\neq j}}\frac{n_s(\varepsilon_j)-n_s(\varepsilon_i)}{\varepsilon_i-\varepsilon_j}\varphi_i(\mathbf{r}_1)\varphi_i^*(\mathbf{r}_2)\varphi_j^*(\mathbf{r}_1)\varphi_j(\mathbf{r}_2) ,\end{aligned} \tag{A16}$$

where $n_s(\varepsilon_i)$ is the occupation number of the ith level (with s being the spin orientation, "up" or "down"):

$$n_s(\varepsilon_i)=\Theta(\varepsilon_F-\varepsilon_i)\equiv\begin{cases}1, & \varepsilon_i\leq\varepsilon_F\\ 0, & \varepsilon_i>\varepsilon_F .\end{cases} \tag{A17}$$

ε_F is the highest occupied level. An analogous, but lengthier expression can also be given for $\chi_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[u])$ in terms of the quantities ε_i, $n_s(\varepsilon_i)$, and $\varphi_i(\mathbf{r})$.

Next, we require the perturbation series for the chemical potential $\mu[u]$ defined by (6). Using (A2) we have

$$\begin{aligned}\mu(u+\Delta u)=\mu[u]+&\int d\mathbf{r}\frac{\partial\rho(\mathbf{r}_i[u])}{\partial N}\Delta u(\mathbf{r})-\tfrac{1}{2}\int d\mathbf{r}_1 d\mathbf{r}_2\frac{\partial}{\partial N}\chi_2(\mathbf{r}_1,\mathbf{r}_{2;}[u])\Delta u(\mathbf{r}_1)\Delta u(\mathbf{r}_2)\\ &-\frac{1}{3!}\int d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3\frac{\partial}{\partial N}\chi_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[u])\Delta u(1)\Delta u(2)\Delta u(3)-\cdots .\end{aligned} \tag{A18}$$

From (A18) we obtain the functional derivatives

$$\left.\frac{\delta\mu[u]}{\delta u(\mathbf{r})}\right|_N=\left.\frac{\partial\rho(\mathbf{r};[u])}{\partial N}\right|_u>0 , \tag{A19}$$

$$\left.\frac{\delta^2\mu[u]}{\delta u(1)\delta u(2)}\right|_N=-\left.\frac{\partial}{\partial N}\chi_2(\mathbf{r}_1,\mathbf{r}_2;[u])\right|_u , \tag{A20}$$

where the postulated inequality in (A19) is a consequence of the fact that $\rho(\mathbf{r})$ is an additive, i.e., monotonically increasing function of N.

We are now in a position to relate the quantities $\bar{\chi}(\mathbf{r}_1,\mathbf{r}_2,\ldots;[\bar{u}])$ defined by Eqs. (31) and (32) to the above response functions $\chi(\mathbf{r}_1,\mathbf{r}_2,\ldots;[u])$. Because the latter are functional derivatives at fixed N, we use the relation

$$\left.\frac{\delta}{\delta u(\mathbf{r})}\right|_N=\frac{\delta}{\delta\bar{u}(\mathbf{r})}-\left.\frac{\delta\mu[u]}{\delta u(\mathbf{r})}\right|_N\int d\mathbf{r}'\frac{\delta}{\delta\bar{u}(r')} , \tag{A21}$$

which follows from the defining relation $\bar{u}(\mathbf{r})\equiv u(\mathbf{r})-\mu$. Applying (A21) first to the density $\rho(\mathbf{r})$, and recalling relations (29), (31), (A3), and (A4), we have

$$\chi_2(\mathbf{r},\mathbf{r}';[u])=\bar{\chi}_2(\mathbf{r},\mathbf{r}';[\bar{u}])-\left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}')}\right|_N\int d\mathbf{r}''\bar{\chi}_2(\mathbf{r},\mathbf{r}'';[\bar{u}]) . \tag{A22}$$

Using Eq. (A11), we obtain a condition on the function $\bar{\chi}_2$:

$$0=\int d\mathbf{r}'\bar{\chi}_2(\mathbf{r},\mathbf{r}';[\bar{u}])-\left.\frac{\delta\mu[u]}{\delta u(\mathbf{r})}\right|_N\int\int d\mathbf{r}'d\mathbf{r}''\bar{\chi}_2(\mathbf{r}',\mathbf{r}'';[\bar{u}]) . \tag{A23}$$

Using Eqs. (36) and (38), (A23) can be solved for $\delta\mu[u]/\delta u(\mathbf{r})|_N$:

$$\left.\frac{\delta\mu[u]}{\delta u(\mathbf{r})}\right|_N=\left.\frac{\partial\rho(\mathbf{r})}{\partial\mu}\right|_u\frac{\partial\mu[u]}{\partial N} , \tag{A24}$$

with the property

$$\int d\mathbf{r}\left.\frac{\delta\mu[u]}{\delta u(\mathbf{r})}\right|_N = 1 \;. \tag{A25}$$

Equations (A24) and (A25) also follow from Eq. (A19); however, their derivation via Eqs. (A21)–(A23) and (A11) provides a check on the consistency of the analysis. With Eqs. (38) and (A24), we can rewrite the general relation between χ_2 and $\bar{\chi}_2$, Eq. (A22), in the form

$$\chi_2(\mathbf{r},\mathbf{r}';[u]) = \bar{\chi}_2(\mathbf{r},\mathbf{r}';[\bar{u}]) - \frac{\partial\mu[u]}{\partial N}\left[\frac{\partial\rho(\mathbf{r})}{\partial\mu}\right]_u \left[\frac{\partial\rho(\mathbf{r}')}{\partial\mu}\right]_u \;. \tag{A26}$$

This equation, together with Eqs. (9), (11), and (33), establish the connection of the two-point correlation function $w_2(\mathbf{r},\mathbf{r}';[\rho])$ and the N-particle linear response function $\chi_2(\mathbf{r},\mathbf{r}';[u])$ for noninteracting fermions in the potential $u(\mathbf{r})$.

Next, we wish to establish the relation between the functions χ_3 and $\bar{\chi}_3$. Applying the operation (A21) to (A22), we obtain

$$\begin{aligned}\chi_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[u]) = {}& \bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\bar{u}]) - \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_2)}\right|_N \int d\mathbf{r}_4\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_3,\mathbf{r}_4;[u])\\ & - \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_3)}\right|_N \int d\mathbf{r}_4\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_3,\mathbf{r}_4;[\bar{u}]) - \left.\frac{\delta^2\mu[u]}{\delta u(\mathbf{r}_2)\delta u(\mathbf{r}_3)}\right|_N \int d\mathbf{r}_2\bar{\chi}_2(\mathbf{r}_1,\mathbf{r}_2;[\bar{u}])\\ & + \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_2)}\right|_N \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_3)}\right|_N \int d\mathbf{r}_2 d\mathbf{r}_3\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\bar{u}]) \;.\end{aligned} \tag{A27}$$

Integrating (A27) with respect to $\mathbf{r}_1$ and using the property (A12) we obtain

$$\begin{aligned}\int d\mathbf{r}_1\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\bar{u}]) = {}& \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_2)}\right|_N \int d\mathbf{r}_1 d\mathbf{r}_4\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_3,\mathbf{r}_4;[\bar{u}]) + \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_3)}\right|_N \int d\mathbf{r}_1 d\mathbf{r}_4\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_4);[\bar{u}]\\ & + \left.\frac{\delta^2\mu[u]}{\delta u(\mathbf{r}_2)\delta u(\mathbf{r}_3)}\right|_N \int d\mathbf{r}_1 d\mathbf{r}_2\bar{\chi}_2(\mathbf{r}_1,\mathbf{r}_2;[\bar{u}]) - \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_2)}\right|_N \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_3)}\right|_N \int d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\bar{u}]) \;,\end{aligned} \tag{A28}$$

which can be simplified with the aid of the relation (38), and the relation derived from it by differentiation with respect to μ, to read

$$\begin{aligned}\int d\mathbf{r}_1\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\bar{u}]) = {}& -\left[\left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_2)}\right|_N \left.\frac{\partial^2\rho(\mathbf{r}_3)}{\partial\mu^2}\right|_u + \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_3)}\right|_N \left.\frac{\partial^2\rho(\mathbf{r}_2)}{\partial\mu^2}\right|_u\right]\\ & + \left.\frac{\delta^2\mu[u]}{\delta u(\mathbf{r}_2)\delta u(\mathbf{r}_3)}\right|_N \left.\frac{\partial N}{\partial\mu}\right|_u + \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_2)}\right|_N \left.\frac{\delta\mu[u]}{\delta u(\mathbf{r}_3)}\right|_N \left.\frac{\partial^2 N}{\partial\mu^2}\right|_u \end{aligned} \tag{A29a}$$

$$\begin{aligned} = {}& -\left.\frac{\partial\mu[u]}{\partial N}\right|_u \frac{\partial}{\partial\mu}\left\{\left.\frac{\partial\rho(\mathbf{r}_2)}{\partial\mu}\frac{\partial\rho(\mathbf{r}_3)}{\partial\mu}\right|_u\right\} - \left.\frac{\partial N}{\partial\mu}\right|_u \left.\frac{\partial}{\partial N}\chi_2(\mathbf{r}_2,\mathbf{r}_3;[u])\right|_u\\ & + \left.\frac{\partial^2 N}{\partial\mu^2}\right|_u \left[\left.\frac{\partial\mu[u]^2}{\partial N}\right|_u\right]^2 \left.\frac{\partial\rho(\mathbf{r}_2)}{\partial\mu}\frac{\partial\rho(\mathbf{r}_3)}{\partial\mu}\right|_u \;,\end{aligned} \tag{A29b}$$

where, in going from (A29a) to (A29b) we used (A20) and (A24).

With the aid of Eq. (A29b), Eq. (A27) assumes the form

$$\begin{aligned}\chi_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,[u]) = {}& \bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;[\bar{u}]) - 2\frac{\partial^2 N}{\partial\mu^2}\left[\frac{\partial\mu}{\partial N}\right]^3 \left.\frac{\partial\rho(\mathbf{r}_1)}{\partial\mu}\frac{\partial\rho(\mathbf{r}_2)}{\partial\mu}\frac{\partial\rho(\mathbf{r}_3)}{\partial\mu}\right|_u + \left[\frac{\partial\mu}{\partial N}\right]^2 \frac{\partial}{\partial\mu}\left[\left.\frac{\partial\rho(\mathbf{r}_1)}{\partial\mu}\frac{\partial\rho(\mathbf{r}_2)}{\partial\mu}\frac{\partial\rho(\mathbf{r}_3)}{\partial\mu}\right|_u\right]\\ & + \left[\frac{\partial\rho(\mathbf{r}_1)}{\partial\mu}\frac{\partial}{\partial N}\chi_2(\mathbf{r}_2,\mathbf{r}_3;[u]) + \frac{\partial\rho(\mathbf{r}_2)}{\partial\mu}\frac{\partial}{\partial N}\chi_2(\mathbf{r}_1,\mathbf{r}_3;[u]) + \frac{\partial\rho(\mathbf{r}_3)}{\partial\mu}\frac{\partial}{\partial N}\chi_2(\mathbf{r}_1,\mathbf{r}_2;[u])\right]_u \;.\end{aligned} \tag{A30}$$

All the above general relations for an inhomogeneous system simplify considerably in the case of homogeneous gas, as shown in Appendix B.

APPENDIX B: IDENTITIES PERTAINING TO THE UNIFORM GAS

Here we apply some of the relations derived in Appendix A to the uniform Fermi gas considered in Sec. VI. In this way, we shall establish the identity (73) required for the proof of Eq. (70).

The relations for the homogeneous gas are obtained by setting $u(\mathbf{r})=0$ after the functional differentiations called for in Appendix A have been carried out and then invoking spatial invariance. Thus, with $\rho(\mathbf{r})=\rho=N/V$, Eqs. (A19) and (A24) become

$$\left.\frac{\delta\mu[u]}{\delta u(\mathbf{r})}\right|_{N;u=0}=\frac{1}{V}\ , \tag{B1}$$

and (A20) becomes

$$\begin{aligned}\left.\frac{\delta^2\mu[u]}{\delta u(\mathbf{r}_1)\delta u(\mathbf{r}_2)}\right|_{N;u=0}&=-\frac{\partial}{\partial N}\chi_2(\mathbf{r}_1,\mathbf{r}_2;\rho)\\&=-\frac{1}{V}\frac{\partial}{\partial\rho}\chi_2(\mathbf{r}_1,\mathbf{r}_2;\rho)\end{aligned} \tag{B2}$$

where we have explicitly indicated the density dependence of the linear response function χ_2 of the uniform system. Using Eqs. (B1) and (41), (A22) becomes

$$\chi_2(\mathbf{r}_1,\mathbf{r}_2;\rho)=\bar{\chi}_2(\mathbf{r}_1,\mathbf{r}_2;\rho)-\frac{1}{V}\frac{\partial\rho}{\partial\mu}\ , \tag{B3}$$

where $\bar{\chi}_2(\mathbf{r}_1,\mathbf{r}_2;\rho)$ is the function whose Fourier transform $\bar{\chi}(k;\rho)$ is given by (54) and (55). Thus, for the uniform gas, the functions χ_2 and $\bar{\chi}_2$ differ only by a spatially independent quantity, implying that their Fourier transforms are related by

$$\chi(k;\rho)=\bar{\chi}(k;\rho)-\delta_{\mathbf{k},0}\frac{\partial\rho}{\partial\mu}\ , \tag{B4}$$

showing that $\chi(k;\rho)$ and $\bar{\chi}(k;\rho)$ are identical, except at $\mathbf{k}=0$; moreover,

$$\lim_{k\to0}\chi(k;\rho)=\lim_{k\to0}\bar{\chi}(k;\rho)=\frac{\partial\rho}{\partial\mu}\ . \tag{B5}$$

With the aid of Eqs. (B1) and (B2), we can now give Eqs. (A29) and (A30) for the uniform gas

$$\int_V d\mathbf{r}_1\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;\rho)=-\frac{1}{V}\frac{\partial^2\rho}{\partial\mu^2}-\frac{\partial\rho}{\partial\mu}\frac{\partial}{\partial\rho}\chi_2(\mathbf{r}_2,\mathbf{r}_3;\rho)\ , \tag{B6}$$

$$\chi_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;\rho)=\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;\rho)+\frac{1}{V^2}\frac{\partial^2\rho}{\partial\mu^2}+\frac{1}{V}\frac{\partial\rho}{\partial\mu}\frac{\partial}{\partial\rho}[\chi_2(\mathbf{r}_1,\mathbf{r}_2;\rho)+\chi_2(\mathbf{r}_2,\mathbf{r}_3;\rho)+\chi_2(\mathbf{r}_3,\mathbf{r}_1;\rho)]\ . \tag{B7}$$

In (B6), the integration is restricted to the volume of the system. It is easily verified, using Eqs. (A11) and (A12), with $u=0$, that (B7) is consistent with (B6).

With Eq. (B6) we are now in a position to prove identity (73). The required quantity, $\bar{\chi}_3(\mathbf{k},-\mathbf{k},0;\rho)$, is defined by

$$\bar{\chi}_3(\mathbf{k},-\mathbf{k},0;\rho)=\int_V d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3 e^{-i\mathbf{k}\cdot(\mathbf{r}_1-\mathbf{r}_2)}\bar{\chi}_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;\rho)\ . \tag{B8}$$

Using result (B6) and the symmetry of $\bar{\chi}_3(1,2,3;\rho)$ with respect to its arguments, (B8) becomes

$$\begin{aligned}\bar{\chi}_3(\mathbf{k},-\mathbf{k},0;\rho)&=-V\frac{\partial^2\rho}{\partial\mu^2}\delta_{\mathbf{k},0}-\frac{\partial\rho}{\partial\mu}\int_V d\mathbf{r}_1 d\mathbf{r}_2 e^{-i\mathbf{k}\cdot(\mathbf{r}_1-\mathbf{r}_2)}\frac{\partial}{\partial\rho}\chi_2(\mathbf{r}_1,\mathbf{r}_2;\rho)\\&=-V\frac{\partial^2\rho}{\partial\mu^2}\delta_{\mathbf{k},0}-V\frac{\partial\rho}{\partial\mu}\frac{\partial}{\partial\rho}\chi(k;\rho)\ .\end{aligned} \tag{B9}$$

Hence, for $\mathbf{k}\neq\mathbf{0}$, with (59) and (B5),

$$\begin{aligned}\frac{\rho d}{V}\bar{\chi}_3(\mathbf{k},-\mathbf{k},0;\rho)&=-\frac{\partial\rho}{\partial\mu}\rho d\frac{\partial}{\partial\rho}\bar{\chi}(k;\rho)\\&=-\bar{\chi}(k\to0;\rho)\rho d\frac{\partial}{\partial\rho}\bar{\chi}(k;\rho)\ ,\end{aligned} \tag{B10}$$

and thus identity (73) is established.

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