Linear-response functions in spin-density-functional theory

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A model system is studied for which exact results are known in order to exhibit directly the limits of density-functional theory when applied to excitation properties. As one might intuitively expect, the ground-state theory accurately describes properties reflecting certain static limits, the paramagnetic susceptibility, for example. The ground-state theory does not, however, accurately describe physical properties involving the density of low-lying excitations, spin-wave, and plasmon dispersion, for example. To demonstrate these limitations we have developed a spin-density-functional theory of the frequency and wave-vector-dependent linear response of the inhomogeneous magnetic electron gas. It is shown that in the local-exchange approximation of Gaspar-Kohn-Sham, the static long-wavelength limits of these functions are identical to those obtained from the time-dependent Hartree-Fock theory [random-phase approximation (RPA) with exchange contributions] of the uniform magnetic electron system in the same limits; however, the plasmon dispersion relation $\omega_{\rm pl}^2(q) \cong \omega_{\rm pl}^2(0)[1 + A(\zeta)(q/k_F)^2]$, and the spin-wave dispersion $\omega_{\rm sw}(q) \cong D(\zeta)(q/k_F)^2$ for long wavelengths $q/k_F \ll 1$, do not correspond with the RPA results. In fact, we find, $A_{\rm LD}(\zeta) < A_{\rm RPA}(\zeta)$ and $D_{\rm LD}(\zeta) > D_{\rm RPA}(\zeta)$, where ζ is the magnetization of the system. The discrepancies found in the dynamical limit are a reflection of the fact that the low-lying excitations close to the ground state are not described by the local scheme. Some comments are made as to the modifications needed in the time-dependent spin-density-functional theory, which may rectify these discrepancies at least in the low-frequency long-wavelength limit.

I. INTRODUCTION

In a series of three papers Hohenberg and Kohn,[1] Kohn and Sham,[2] and Sham and Kohn[3] have laid the foundations for a new theory of electronic structure of nonmagnetic systems. Hedin and Lundquist[4] have made this theory into a practical calculational scheme incorporating the recent advances in the theory of the interacting electron gas. The generalization of this theory to make it applicable to magnetic systems, as well as to systems where the spin polarization plays an important role, was put forward by von Barth and Hedin[5] and Rajagopal and Callaway.[6] The success of this formulation in elucidating various ground-state properties of atoms, molecules, and solids is best illustrated in the recent work or Gunnarson and Lundquist.[7] Much of this work pertains to the properties of the ground state of the system. In Refs. 3 and 4, an examination of the single-particle states within this scheme was made. Vosko and Perdew[8] developed a theory of the paramagnetic spin susceptibility of an inhomogeneous electron gas based on the Hohenberg-Kohn-Sham (HKS) formalism. Janak[9] made a detailed study of this susceptibility and was able to explain the systematics of the $3d$ and $4d$ series of elements. The theory correctly showed which of the elements are magnetic and which were not—with no exception. Given this remarkable success of the theory for the ground-state properties, one wonders if the low-lying excited states, such as spin waves, close to the ground state of the system may not be described by a suitable extension of the above formalism.

In the theory of the interacting electron gas, one already has such a scheme for self-consistently calculating the various correlation functions of the system. In particular, Herring,[10] using a perturbation theory of the Hartree-Fock self-consistent equations due to Peng,[11] calculated the spin-wave dispersion in the long-wavelength limit for a ferromagnetic electron gas. Rajagopal, Brooks, and Ranganathan,[12] using the Green's-function method were able to set up these equations more generally for a wider class of properties, such as the frequency- and wave-vector-dependent spin susceptibilities and dielectric function of the magnetic electron gas. Rajagopal[13] solved the relevant integral equation pertaining to the transverse spin susceptibility in the low-frequency long-wavelength limit for all ground-state magnetizations and recovered the Herring result in the saturated ferromagnetic case. This variational calculation has been extended to calculate all the susceptibilities by Rajagopal, Rath, and Kimball.[14] In the present paper, we shall employ the same method as given in Ref. 12 to calculate the correlation functions for the inhomogeneous systems in the spin-density-functional formalism. This generalizes the work of Vosko and Perdew[8] to the magnetic case.

Recently, Callaway and Wang[15] have derived an expression for the transverse magnetic susceptibility in the local-exchange approximation of Gaspar-Kohn-Sham. The change in the local-exchange potential when the external field is applied was *suggested* to be of a special form which was consistent with the requirement that for zero wave vector, the spin-wave energy be zero. Moreover, when the spin-wave energy was computed within this scheme, the stiffness coefficient D in $\omega_{sw}(q) \simeq D(q/k_F)^2$ was found to be larger than the value calculated by Herring[10] and Rajagopal.[13] In the present paper, we *derive* the correct form for the change in the exchange potential, in the spin-density-functional theory. When the ground-state energy of the homogeneous electron gas is assumed to depend on the magnitude of the magnetization vector only, we obtain the form assumed by Calaway and Wang,[15] who used the local-exchange scheme only. From our derivation, it is clear that a local-exchange scheme will not give us a reliable estimate of D.

In order to strengthen this belief, we calculated the dispersion of the plasma frequency in the long-wavelength limit in the homogeneous electron gas from Ref. 4: $\omega_{pl}^2(q) \cong \omega_{pl}^2(0)[1 + A(\zeta)(q/k_F)^2]$. Singhal and Callaway[16] have recently developed an expression for the dielectric function in the local-density theory by an extension of the Callaway-Wang scheme. Our expression for the dielectric function reduces to the Singhal-Callaway expression when the local-exchange approximation is made. We computed the dispersion in the plasmon frequency in the long-wavelength limit and found that A_{LD} is smaller than A_{RPA}.

It should be mentioned that the static long-wavelength limit of the correlation functions for the homogeneous electron gas[14] completely coincides with the corresponding static values based on the local-density theory. This is as expected because the local-density theory corresponds to the Hartree-Fock (HF) theory of the uniform gas as far as the ground-state properties are concerned.

We develop the formalism for computing the linear-response functions in Sec. II. In the same section, we apply it to the case when the ground-state energy of a uniform electron gas is assumed to be a function of the magnitude of the magnetization vector in the ground state, an assumption which is reasonable in view of the homogeneity of the uniform electron gas. In Sec. III, we deduce the various limiting values in a local-density scheme and compare with the corresponding results for the uniform gas computed by using RPA with exchange contributions properly taken into account (sometimes known as the time-dependent Hartree-Fock theory). In Sec. IV, we briefly summarize the results and speculate on how the theory may be modified further to at least yield the correct limits.

II. FORMULATION OF THE THEORY

A. Preliminaries

We follow Ref. 12 and introduce the 2×2 matrix Green's function

$$\underline{G}_{\sigma\sigma'}(1;1') = (-i)\langle T[\psi_\sigma(1)\psi_{\sigma'}^\dagger(1')]\rangle \ , \tag{1}$$

where $\psi_\sigma(1)$, $\psi_{\sigma'}^\dagger(1')$ are the destruction and creation operators for an electron at 1 ($\equiv \vec{r}_1, t_1$) with spin σ and at $1'(\equiv \vec{r}_1', t_1')$ with spin σ'. These operators obey the usual anticommutation rules at equal times. The symbol T here denotes the time ordering as usual and $\underline{G}_{\sigma\sigma'}$ then obeys an antiperiodic condition in the complex time domain[17] and $\langle \cdots \rangle$ signifies the grand canonical ensemble average. This Green's function obeys the Dyson equation

$$\left[\left(i\frac{\partial}{\partial t_1} + \frac{\nabla_1^2}{2m} - V(r_1) - U_0(1)\right)\underline{\tau}_0 - \vec{U}(1)\cdot\vec{\underline{\tau}}\right]\underline{G}(11') - \int \underline{\Sigma}(1\bar{1})\underline{G}(\bar{1}1')\,d\bar{1} = \underline{\tau}_0\delta(11') \ . \tag{2}$$

Here $\underline{\Sigma}$ is the so-called mass operator and is also a 2×2 matrix. It is in general complex. We introduce the standard Pauli spin matrices:

$$\underline{\tau}_0 = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \ , \quad \underline{\tau}_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \ , \qquad \underline{\tau}_2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \ , \quad \underline{\tau}_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \ . \tag{3}$$

We use units where $\hbar = 1$. The time integration in Eq. (2) is from 0 to $-i\beta$, $\beta = 1/k_B T$, in the standard manner. In Eq. (2), $V(r_1)$ is the local potential, due to, say, the ions; $U_0(\vec{r}_1 t_1)$ is the external time-dependent potential and $\vec{U}(\vec{r}_1 t_1)$ is the external time-dependent magnetic field in appropriate units. (We use units where the Bohr magneton is unity.) It is now clear from (1) that the average particle density and the spin-density vector are given by[12]

$$n(1) = -i\,\mathrm{Tr}[\underline{\tau}_0\underline{G}(11^+)] \equiv n_0(1) \tag{4}$$

and

$$\vec{s}(1) = -i\,\mathrm{Tr}[\vec{\underline{\tau}}\underline{G}(11^+)] \equiv (n_1(1), n_2(1), n_3(1)) \ , \tag{5}$$

where Tr is the trace over the spin indices. Often we denote the components of the vector by a greek index α. It may also be pointed out that the inverse of the Green's function $\underline{G}^{-1}$ is defined by

$$\int \underline{G}^{-1}(1\bar{1})\,\underline{G}(\bar{1}1')\,d\bar{1} = \underline{\tau}_0\delta(11') = \int \underline{G}(1\bar{1})\underline{G}^{-1}(\bar{1}1')\,d\bar{1} \ . \tag{6}$$

The linear-response functions are defined by

$$\chi_{ij}(12) \equiv (-i)\langle T[\hat{n}_i(1)\hat{n}_j(2)]\rangle \ , \tag{7}$$

where

$$\hat{n}_i(1) = n_i^{\rm op} - n_i, \quad n_i^{\rm op} = {\rm Tr}(\underline{\Psi}^\dagger \underline{\tau}_i \underline{\Psi})$$

is the operator, Ψ is the column vector $(\psi_\uparrow, \psi_\downarrow)$, and $\Psi^\dagger$ its transpose. Expressions (7) may be reexpressed in terms of $\underline{G}$ and its variational derivatives[12]

$$\chi_{ij}(12) \equiv \frac{\delta n_i(1)}{\delta U_j(2)} = -i{\rm Tr}\left[\underline{\tau}_i \frac{\delta G(11^+)}{\delta U_j(2)}\right]$$

or

$$\chi_{ij}(12) = i\int {\rm Tr}[\underline{\tau}_i \underline{G}(1\bar{1}) \times \underline{\Gamma}_j(\bar{1}\bar{2};2)\underline{G}(\bar{2}1^+)]\,d\bar{1}\,d\bar{2} \ , \quad (8)$$

where we have made use of Eq. (6) in introducing the vertex function

$$\underline{\Gamma}_i(12;3) = \delta\underline{G}^{-1}(12)/\delta U_i(3) \ . \quad (9)$$

From Eq. (2), $\underline{G}^{-1}(12)$ is given by

$$\underline{G}^{-1}(12) = \left[\left(i\frac{\partial}{\partial t_1} + \frac{\nabla_1^2}{2m} - V(r_1) - U_0(1)\right) \times \underline{\tau}_0 - \vec{\tau}\cdot\vec{U}(1)\right]\delta(12) - \underline{\Sigma}(12) \ . \quad (10)$$

Once $\underline{\Sigma}(12)$ is known in some approximation, Γ_i can be computed and hence the χ_{ij}'s can be determined. This then is our program.

B. Spin-density-functional approximation for $\underline{\Sigma}(12)$ in the absence of U_i

An argument similar to the one given in Refs. 3 and 4 can now be constructed for the spin-density-functional version for the self-energy $\underline{\Sigma}$. For the equilibrium state,[18] $\underline{\Sigma}(12)$ is a functional of n, $\vec{s}$, and in fact within a static approximation, we may write

$$\underline{\Sigma}(12) \cong (\underline{\tau}_0 V_{xc}[\vec{r}_1, n, \vec{s}] + \underline{\tau}_0 V_H[\vec{r}_1, n] + \vec{\tau}\cdot\vec{W}_{xc}[\vec{r}_1, n, \vec{s}])\delta(12) \ , \quad (11)$$

where

$$V_{xc}[\vec{r}_1, n, \vec{s}] \equiv \delta E_{xc}[n, \vec{s}]/\delta n(\vec{r}_1) \ , \quad (12)$$

$$\vec{W}_{xc}[\vec{r}_1, n, \vec{s}] \equiv \delta E_{xc}[n, \vec{s}]/\delta\vec{s}(\vec{r}_1) \ , \quad (13)$$

with $E_{xc}[n, \vec{s}]$ the exchange-correlation part of the ground-state energy of the inhomogeneous many-body system, $V_H[\vec{r}_1, n]$ is the Hartree self-energy

$$V_H[\vec{r}_1, n] = e^2\int \frac{1}{|\vec{r}_1 - \vec{r}_1'|} n(\vec{r}_1')\,d^3r_1' \ . \quad (14)$$

The Green's function in the absence of the external fields U_i is denoted by $\underline{G}_0(12)$ and for the equilibrium state, we may write it in terms of the complete set of orthonormal spinor functions which obey the equations[5,6]

$$E_i^\lambda \phi_{i\sigma_1}^\lambda(\vec{r}_1) = \left(\frac{\nabla_1^2}{2m} + V(\vec{r}_1) + V_{xc}[\vec{r}_1, n, \vec{s}] + V_H[\vec{r}_1, n]\right)\phi_{i\sigma_1}^\lambda(\vec{r}_1) + \sum_{\sigma_1'} \vec{W}_{xc}[\vec{r}_1, n, \vec{s}]\cdot\vec{\tau}_{\sigma_1\sigma_1'}\phi_{i\sigma_1'}^\lambda(\vec{r}_1) \quad (\lambda = 1, 2) \ . \quad (15)$$

Here i labels the appropriate quantum numbers and λ stands for the eigenspinor. The Green's function $\underline{G}_0(12)$ is then

$$\underline{G}_0(12) = \frac{1}{-i\beta}\sum_n \sum_{i,\lambda=1,2} e^{iE_n(t_1 - t_2)} g_i^\lambda(E_n) \times \begin{pmatrix} \phi_{i\uparrow}^\lambda(\vec{r}_1) \\ \phi_{i\downarrow}^\lambda(\vec{r}_1) \end{pmatrix} \times [\phi_{i\uparrow}^{(\lambda)*}(r_2)\phi_{i\downarrow}^{(\lambda)*}(r_2)] \ , \quad (16)$$

with $E_n = (2n+1)\pi/(-i\beta) + \mu$, μ being the chemical potential, $n = 0, \pm1, \pm2, \ldots$, and

$$g_i^\lambda(E_n) = 1/(E_n - E_i^\lambda) \ . \quad (17)$$

The various densities are then computed in the usual way and we have

$$n_0(\vec{r}) = \sum_{i\lambda}\sum_\sigma |\phi_{i\sigma}^\lambda(\vec{r})|^2 n_F(E_i^\lambda) \ , \quad (18)$$

$$S_{z0}(\vec{r}) = \sum_{i\lambda}(|\phi_{i\uparrow}^\lambda(\vec{r})|^2 - |\phi_{i\downarrow}^\lambda(\vec{r})|^2) n_F(E_i^\lambda) \ , \quad (19)$$

$$S_{+0}(\vec{r}) = \sum_{i\lambda}\phi_{i\downarrow}^\lambda(\vec{r})\phi_{i\uparrow}^{\lambda*}(\vec{r}) n_F(E_i^\lambda) \ , \quad (20)$$

$$S_{-0}(\vec{r}) = \sum_{i\lambda}\phi_{i\uparrow}^\lambda(\vec{r})\phi_{i\downarrow}^{\lambda*}(\vec{r}) n_F(E_i^\lambda) \ . \quad (21)$$

Here $n_F(E_i^\lambda)$ is the usual Fermi function

$$n_F(E_i^\lambda) = 1/[\exp\beta(E_i^\lambda - E_F^\lambda) + 1] \ ,$$

where E_F^λ are the two Fermi energies, determined from the condition that the total number and the total magnetization vector are given constants. The subscripts 0 in Eqs. (18)–(21) denote that these are appropriate to the ground state. The functions $\phi_{i\sigma}^\lambda(\vec{r})$ obey the usual orthonormal relationships:

$$\int [\phi_{i\uparrow}^{\lambda*}(\vec{r})\phi_{i\downarrow}^{\lambda'*}(\vec{r})] \begin{pmatrix} \phi_{j\uparrow}^{\lambda'}(\vec{r}) \\ \phi_{j\downarrow}^{\lambda'}(\vec{r}) \end{pmatrix} d^3r = \delta_{ij}\delta_{\lambda\lambda'} \quad (22a)$$

and

$$\sum_{i\lambda}\phi_{i\sigma}^\lambda(\vec{r})\phi_{i\sigma'}^{\lambda*}(\vec{r}') = \delta_{\sigma\sigma'}\delta(\vec{r} - \vec{r}') \ . \quad (22b)$$

Note that Eqs. (15) and (18)–(21) form the self-consistent set, to be solved eventually. These reduce to the form given by Vosko and Perdew in the paramagnetic limit.

C. Derivation of χ_{ij}

We now assume that the external time-dependent fields $U_i(\vec{r}t)\,(i=0,1,2,3)$ are *slowly* turned on so that the equilibrium state follows the external field in an equally slow way. This implies that we compute the changes induced in n, $\vec{s}$ by U_i to first order from Eqs. (4), (5), and (10) with $\underline{\Sigma}$ given by Eq. (11):

$$n_i(1) = n_{i0}(\vec{r}_1) + i\sum_{k=0}^{3}\int \mathrm{Tr}[\underline{\tau}_i\underline{G}_0(1\bar{1})\underline{\Gamma}_k(\bar{1}\bar{2};\bar{3})\underline{G}_0(\bar{2}1^+)] \times U_k(\bar{3})\,d\bar{1}\,d\bar{2}\,d\bar{3} \quad (j=0,1,2,3)\ . \tag{23}$$

Now, $\underline{\Gamma}_k(12;3)$ is obtained from Eq. (10) with Eq. (11) for $\underline{\Sigma}$,

$$\underline{\Gamma}_k(12;3) = -\underline{\tau}_k\delta(12)\delta(13) - \delta(12)\underline{\tau}_0\left[\int [V_c(1\bar{1}) + K_{xc}(1\bar{1})]\chi_{0k}(\bar{1}3)\,d\bar{T} + \sum_{\alpha=1}^{3}\int G^{(1)}_{xc\,\alpha}(1\bar{1})]\chi_{\alpha k}(\bar{1}3)\,d\bar{T}\right]$$

$$-\delta(12)\left[\sum_{\alpha=1}^{3}\int \underline{\tau}_\alpha G^{(1)}_{xc\,\alpha}(1\bar{1})\chi_{0k}(\bar{1}3)\,d\bar{1} + \sum_{\alpha,\beta=1}^{3}\int \underline{\tau}_\alpha G^{(2)}_{xc\,\alpha\beta}(1\bar{1})\chi_{\beta k}(\bar{1}3)\,d\bar{1}\right]\ . \tag{24}$$

Here we have introduced the notations

$$V_c(12) = V_c(|\vec{r}_1-\vec{r}_2|)\delta(t_1-t_2)\ , \tag{25a}$$

where the V_c stands for instantaneous Coulomb potential;

$$K_{xc}(12) = \frac{\delta^2 E_{xc}}{\delta n(\vec{r}_1)\delta n(\vec{r}_2)}\delta(t_1-t_2)\ , \tag{25b}$$

$$G^{(1)}_{xc\,\alpha}(12) = \frac{\delta^2 E_{xc}}{\delta n(\vec{r}_1)\delta s_\alpha(\vec{r}_2)}\delta(t_1-t_2) = \frac{\delta^2 E_{xc}}{\delta s_\alpha(\vec{r}_2)\delta n(\vec{r}_1)}\delta(t_1-t_2)\ , \tag{25c}$$

$$G^{(2)}_{xc\,\alpha\beta}(12) = \frac{\delta^2 E_{xc}}{\delta s_\alpha(\vec{r}_1)\delta s_\beta(\vec{r}_2)}\delta(t_1-t_2)\ . \tag{25d}$$

Thus the susceptibilities are determined from the set of linear equations using the definition (8) and Eq. (24):

$$\chi_{ij}(12) = \chi^{(0)}_{ij}(12) + \int \chi^{(0)}_{i0}(1\bar{1})[V_c(\bar{1}\bar{2}) + K_{xc}(\bar{1}\bar{2})\chi_{0j}(\bar{2}2)\,d\bar{1}\,d\bar{2} + \int \chi^{(0)}_{i0}(1\bar{1})\sum_{\alpha=1}^{3}G^{(1)}_{xc\,\alpha}(\bar{1}\bar{2})\chi_{\alpha j}(\bar{2}2)\,d\bar{1}\,d\bar{2}$$

$$+\int\sum_{\alpha=1}^{3}\chi^{(0)}_{i\alpha}(1\bar{1})G^{(1)}_{xc\,\alpha}(\bar{1}\bar{2})\chi_{0j}(\bar{2}2)\,d\bar{1}\,d\bar{2} + \int\sum_{\alpha,\beta=1}^{3}\chi^{(0)}_{i\alpha}(1\bar{1})G^{(2)}_{xc\,\alpha\beta}(\bar{1}\bar{2})\chi_{\beta j}(\bar{2}2)\,d\bar{1}\,d\bar{2}\ . \tag{26}$$

Here we have introduced the "noninteracting" correlation functions

$$\chi^{(0)}_{ij}(12) = -i\,\mathrm{Tr}[\underline{\tau}_i\underline{G}_0(12)\underline{\tau}_j\underline{G}_0(21^+)]\ . \tag{27}$$

These equations together with Eq. (16) for $\underline{G}_0$ generalize the Vosko-Perdew equations for the magnetic case.

D. Simplified model

A neat reduction in the number of functions of χ_{ij} can be achieved if a model for the magnetic system can be made. The most reasonable assumption that may be made stems from our knowledge of the homogeneous electron gas. Consider a single domain fer-

romagnet. Then the magnetization vector in the absence of external fields may be assumed to be directed along a fixed direction. Then we can assume, in view of the time-reversal symmetry, that the exchange-correlation energy $E_{xc}[n,\vec{s}_0]$ is a function of the magnitude of $|\vec{s}_0|$ only:

$$E_{xc}[n,\vec{s}_0] \equiv E_{xc}[n,|\vec{s}_0|] \ . \tag{28}$$

Also, we may choose this direction of magnetization to be the Z axis. Then

$$W_{xc\,\alpha}[\vec{r}_1,n,\vec{s}_0] = W_{xc}[\vec{r}_1,n,|\vec{s}_0|]s_{\alpha 0}(\vec{r}_1)/|\vec{s}_0(\vec{r}_1)|$$

or

$$\vec{W}_{xc}[\vec{r}_1,n,\vec{s}_0] = W_{xc}[\vec{r}_1,n,|\vec{s}_0|](0,0,1) \ , \tag{29}$$

where

$$W_{xc}[\vec{r}_1,n,|\vec{s}_0|] = \delta E_{xc}[n,|\vec{s}_0|]/\delta|\vec{s}_0(\vec{r}_1)| \ . \tag{30}$$

Hence $\underline{G}_0(12)$ takes a very simple form, because the above assumption implies that there exists a "principal-axis" transformation of the spinor wave functions $\{\phi^{\lambda}_{i\sigma}(\vec{r})\}$ which makes $\underline{G}_0$ diagonal

$$\underline{G}_0(12) = \frac{1}{(-i\beta}\sum_n\sum_i \begin{pmatrix} \Psi_{i\uparrow}(\vec{r}_1)\Psi_{i\uparrow}^*(\vec{r}_2)g_{i\uparrow}(E_n) & 0 \\ 0 & \Psi_{i\downarrow}(\vec{r}_1)\Psi_{i\downarrow}^*(\vec{r}_2)g_{i\downarrow}(E_n) \end{pmatrix} , \tag{31}$$

where

$$g_{i\sigma}(E_n) = 1/(E_n - E_{i\sigma}) \ , \tag{32}$$

and the $\{\Psi_{i\sigma}(\vec{r})\}$ are the solutions of the equations

$$E_{i\sigma}\Psi_{i\sigma}(\vec{r}) = \left(-\frac{\nabla^2}{2m} + V(\vec{r}) + V_{xc}[\vec{r},n,|\vec{s}_0|] + V_H[\vec{r},n]\right)\Psi_{i\sigma}(\vec{r}) + \tau_{3\sigma\sigma}W_{xc}[\vec{r},n,|\vec{s}_0|]\Psi_{i\sigma}(\vec{r}) \ . \tag{33}$$

With this form for $\underline{G}_0(12)$, we at once obtain a tremendous reduction in the number of nonzero $\chi^{(0)}_{\mu\mu}$. In fact, we have only the following nonzero $\chi^{(0)}_{\mu\mu}$'s:

$$\chi^{(0)}_{00},\ \chi^{(0)}_{03} = \chi^{(0)}_{30},\ \chi^{(0)}_{33},\ \chi^{(0)}_{+-},\ \text{and}\ \chi^{(0)}_{-+} \ ,$$

with

$$\chi^{(0)}_{+-} = (-i)\langle T(\tfrac{1}{2}(n_1+in_2),\tfrac{1}{2}(n_1-in_2))\rangle \ , \text{etc.} \tag{34}$$

Furthermore, we have

$$\vec{G}^{(1)}_{xc}(12) = G^{(1)}_{xc}(12)(0,0,1) \ , \tag{35}$$

where

$$G^{(1)}_{xc}(12) = \frac{\delta^2 E_{xc}}{\delta n(\vec{r}_1)\delta|\vec{s}_0(\vec{r}_2)|}\delta(t_1-t_2) \ ;$$

and after a little algebra, one finds

$$G^{(2)}_{xc\,\alpha\beta}(12) = 0 \quad \text{for} \quad \alpha \neq \beta \ , \tag{36a}$$

$$G^{(2)}_{xc\,11}(12) = G^{(22)}_{xc\,22}(12) \equiv G^{(2)}_{xc\perp}(12) = \delta(12)\frac{W_{xc}[\vec{r}_1,n,|\vec{s}_0|]}{|\vec{s}(\vec{r}_1)|} \ , \tag{36b}$$

and

$$G^{(2)}_{xc\,33}(12) \equiv G^{(2)}_{xc\,\parallel}(12) = \delta(t_1-t_2) \times \frac{\delta^2 E_{xc}}{\delta|\vec{s}_0(\vec{r}_1)|\delta|\vec{s}_0(\vec{r}_2)|} \ . \tag{36c}$$

We then find the components χ_{00}, $\chi_{03}=\chi_{30}$, χ_{33} decouple from the "transverse" spin susceptibilities χ_{+-}, χ_{-+}. Observe that the particle-density response and the longitudinal spin-response functions are coupled, a result first pointed out in Ref. 12 for the homogeneous system.

We thus obtain

$$\chi_{+-}(12) = \chi^{(0)}_{+-}(12) + 2\int \chi^{(0)}_{+-}(1\bar{1}) \times G^{(2)}_{xc\perp}(\bar{1}\bar{2})\chi_{+-}(\bar{2}2)\,d\bar{1}\,d\bar{2} \ , \tag{37}$$

and a similar equation $\chi_{-+}(12)$. The equations for χ_{00} and χ_{30}, and χ_{33} and χ_{03} are coupled and can be written down similarly, following Eqs. (26), (35), and (36). Equations (26) and (37) are the generalization of the Vosko-Perdew expression for the magnetic case. Using the corresponding expressions for $\chi^{(0)}_{\mu\mu}$ in terms of the "wave functions," (33), we can express $\chi_{\mu\mu}$ in the same way. Instead of giving all these expressions, we shall now make a few comments as to the physical nature of these equations.

In the paramagnetic limit, we have

$$G^{(1)}_{xc} = 0 \ , \tag{38}$$

$$G^{(2)}_{xc\,\parallel} = G^{(2)}_{xc\perp} \ ; \tag{39}$$

also $\chi^{(0)}_{03} = 0 = \chi^{(0)}_{30}$. The equations for χ_{00} and χ_{33} then decouple. Since $\chi_{+-} = 2\chi_{33}$ in this case, the equation for χ_{+-} coincide with that of χ_{33}.

From Eqs. (33), we note that W_{xc} can be interpreted as the potential responsible for the splitting of the spin up and down bands for the electrons. $G^{(1)}_{xc}$ is an effective interaction between the magnetization and

particle density, and comes about because these two quantities arise from the electron densities in the up and down spin configurations. K_{xc} is the usual effective interaction between the particle densities at the two different points in the medium, whereas $G^{(2)}_{xc\parallel}$ is the corresponding interaction between the magnetization densities along the polarization direction of the intrinsic magnetization of the system. But $G^{(2)}_{xc\perp}$ is the interaction between the two transverse components of the induced magnetization; this provides the mechanism for the intrinsic rotation of the spin direction from the equilibrium state magnetization. Notice that unlike the other effective interactions, $G^{(2)}_{xc\perp}$ is *local*. Usually, one further assumes $K_{xc}, G^{(2)}_{xc\parallel}$ to be local, to facilitate computation.[8,9,16] In Sec. III we shall specialize these results for the homogeneous electron gas, using the simplest local form for E_{xc}.

III. APPLICATION TO THE HOMOGENEOUS ELECTRON GAS

A. Local-density scheme ($T=0°$ K)

Detailed analytic expressions can be obtained if we assume E_{xc} to be just the Hartree-Fock energy of the homogeneous gas[12] (units where $e^2=1$ and $\hbar=1$ are employed here)

$$E_{xc} \cong E_x = -\tfrac{3}{8}(3/\pi)^{1/3}[(n+s)^{4/3}+(n-s)^{4/3}] \ . \tag{40}$$

n, s are independent of r, and are given by

$$n = k_F^3/3\pi^2, \quad s = \zeta k_F^3/3\pi^2 \ , \tag{41}$$

with k_F the Fermi momentum of the system and ζ is the intrinsic magnetization of the system and varies between 0 and 1. Then

$$V_{xc} = -\tfrac{1}{2}(3/\pi)^{1/3}[(n+s)^{1/3}+(n-s)^{1/3}] \ , \tag{42a}$$

$$W_{xc} = -\tfrac{1}{2}(3/\pi)^{1/3}[(n+s)^{1/3}-(n-s)^{1/3}] \ , \tag{42b}$$

$$G^{(1)}_{xc} = -\tfrac{1}{6}(3/\pi)^{1/3}[(n+s)^{-2/3}-(n-s)^{-2/3}]\delta(12) \ , \tag{43a}$$

$$K_{xc} = G^{(2)}_{xc\parallel} = -\tfrac{1}{6}(3/\pi)^{1/3}[(n+s)^{-2/3} + (n-s)^{-2/3}]\delta(12) \ , \tag{43b}$$

and

$$G^{(2)}_{xc\perp} = \frac{1}{s}W_{xc} = -\tfrac{1}{2}(3/\pi)^{1/3}\{[(n+s)^{1/3} - (n-s)^{1/3}]/s\}\delta(12) \ . \tag{43c}$$

In this case, plane waves are solutions of Eq. (33), and $\chi^{(0)}_{\sigma\sigma'}$ are given by[12-14]

$$\chi^{(0)}_{00}(q,\omega) = A^{(0)}_{\uparrow\uparrow}(q,\omega) + A^{(0)}_{\downarrow\downarrow}(q,\omega) = \chi^{(0)}_{33}(q,\omega) \ , \tag{44a}$$

$$\chi^{(0)}_{03}(q,\omega) = \chi^{(0)}_{30}(q,\omega) = A^{(0)}_{\uparrow\uparrow}(q,\omega) - A^{(0)}_{\downarrow\downarrow}(q,\omega) \ , \tag{44b}$$

and

$$\chi^{(0)}_{+-}(q,\omega) = A^{(0)}_{\uparrow\uparrow}(q,\omega), \quad \chi^{(0)}_{+-}(q,\omega) = A^{(0)}_{\downarrow\downarrow}(q,\omega) \ , \tag{44c}$$

where

$$A^{(0)}_{\sigma\sigma'}(q,\omega) = -\int \frac{d^3k}{(2\pi)^3} \frac{f_\sigma(\vec{k}+\frac{1}{2}\vec{q}) - f_{\sigma'}(\vec{k}-\frac{1}{2}\vec{q})}{\omega + iE - E_\sigma(\vec{k}+\frac{1}{2}\vec{q}) + E_{\sigma'}(\vec{k}-\frac{1}{2}\vec{q})} \ , \tag{45}$$

with $f_\sigma(k)$ being the Fermi function for electrons of spin σ and

$$E_\sigma(\vec{k}) = |\vec{k}|^2/2m + V_{xc} + \eta_\sigma W_{xc} \ . \tag{46}$$

Here $\eta_\sigma = +1$ for $\sigma = \uparrow$ and -1 for $\sigma = \downarrow$.

Explicit expressions for $A^{(0)}_{\sigma\sigma'}(q,\omega)$ are given in Refs. 12 and 14 as well as some of the relevant limits which will be needed here. Then

$$\chi_{+-}(q,\omega) = \chi^{(0)}_{+-}(q,\omega)/[1-(2W_{xc}/s)\chi^{(0)}_{+-}(q,\omega)] \tag{47}$$

and a similar formula for χ_{-+}. Callaway and Wang[15] obtained this formula; they however postulated $G^{(2)}_{xc\perp}$ to be W_{xc}/s, from the requirement that the spin waves given by

$$1 = G^{(2)}_{xc\perp}\chi^{(0)}_{+-}(q,\omega) \tag{47a}$$

have the property that for $q=0$, ω is zero. We have here derived this result. Using the result for $A^{(0)}_{\uparrow\downarrow}$ in Ref. 12 and the expressions (41), (42b), and (43c) we obtain the spin-wave dispersion to be of the form

$$\omega_{sw} = D_{LD}(\zeta)(q/k_F)^2 \ , \tag{48}$$

where

$$D_{LD}(\zeta) = \frac{k_F^2}{2m\zeta}\left[1 - \frac{\pi}{5\alpha r_s}\left[\frac{(1+\zeta)^{5/3}-(1-\zeta)^{5/3}}{(1+\zeta)^{1/3}-(1-\zeta)^{1/3}}\right]\right] \ . \tag{49}$$

Callaway and Wang[15] derived this result for $\zeta=1$ only. Here r_s is the usual dimensionless electron gas parameter, given by $\alpha r_s = 1/k_F$, α is a numerical constant (=0.521).

The other correlation functions can be obtained

directly by Fourier transforming the equations (26) and after some algebra,

$$\chi_{00}(q,\omega) = (A_{\uparrow\uparrow}^{(0)} + A_{\downarrow\downarrow}^{(0)} - 4G_{xc\parallel}^{(2)} A_{\uparrow\uparrow}^{(0)} A_{\downarrow\downarrow}^{(0)})/\mathfrak{D} \ , \tag{50}$$

$$\chi_{03}(q,\omega) = \chi_{30}(q,\omega) = (A_{\uparrow\uparrow}^{(0)} - A_{\downarrow\downarrow}^{(0)} + 4G_{xc}^{(1)} A_{\uparrow\uparrow}^{(0)} A_{\downarrow\downarrow}^{(0)})/\mathfrak{D} \ , \tag{51}$$

$$\chi_{33}(q,\omega) - [A_{\uparrow\uparrow}^{(0)} + A_{\downarrow\downarrow}^{(0)} - 4(K_{xc} + V_c) A_{\uparrow\uparrow}^{(0)} A_{\downarrow\downarrow}^{(0)}]/\mathfrak{D} \ , \tag{52}$$

where

$$\mathfrak{D} = \{1 - (K_{xc} + V_c + G_{xc\parallel}^{(2)})(A_{\uparrow\uparrow}^{(0)} + A_{\downarrow\downarrow}^{(0)}) - 2G_{xc}^{(1)}(A_{\uparrow\uparrow}^{(0)} - A_{\downarrow\downarrow}^{(0)}) + 4[(K_{xc} + V_c) G_{xc\parallel}^{(2)} - (G_{xc}^{(1)})^2] A_{\uparrow\uparrow}^{(0)} A_{\downarrow\downarrow}^{(0)}\} \ . \tag{53}$$

The longitudinal dielectric constant is given by

$$\epsilon(q,\omega) = [1 + V_c(q)\chi_{00}(q,\omega)]^{-1} \ .$$

Using (50) and (53), we obtain after some algebra,

$$\epsilon(q,\omega) = 1 + V_c(q)(4G_{xc\parallel}^{(2)} A_{\uparrow\uparrow}^{(0)} A_{\downarrow\downarrow}^{(0)} - A_{\uparrow\uparrow}^{(0)} - A_{\downarrow\downarrow}^{(0)})/\mathfrak{D}' \tag{54}$$

where

$$\mathfrak{D}' = \{1 - (K_{xc} + G_{xc\parallel}^{(2)})(A_{\uparrow\uparrow}^{(0)} + A_{\downarrow\downarrow}^{(0)}) - 2G_{xc}^{(1)}(A_{\uparrow\uparrow}^{(0)} - A_{\downarrow\downarrow}^{(0)}) + 4[K_{xc} G_{xc\parallel}^{(2)} - (G_{xc}^{(1)})^2] A_{\uparrow\uparrow}^{(0)} A_{\downarrow\downarrow}^{(0)}\} \ . \tag{55}$$

The result that $\chi_{03} = \chi_{30}$ was proved in Ref. 14 to be true in general, in the absence of spin-orbit coupling, and is borne out in the present scheme also. In the paramagnetic limit, the expressions reduce enormously and we obtain

$$A_{\uparrow\uparrow}^{(0)} = A_{\downarrow\downarrow}^{(0)} = A^{(0)}, \quad G_{xc}^{(1)} = 0 \ ,$$

so that

$$\chi_{33}^{P}(q,\omega) = 2A^{(0)}/[1 - 2G_{xc\parallel}^{(2)} A^{(0)}] \ , \tag{56a}$$

$$\chi_{30}^{P}(q,\omega) = 0 = \chi_{03}^{P}(q,\omega) \ , \tag{56b}$$

$$\chi_{00}^{P}(q,\omega) = 2A^{(0)}/[1 - 2(V_c + K_{xc})A^{(0)}] \ , \tag{56c}$$

and

$$\epsilon^{P}(q,\omega) = 1 - 2V_c(q)A^{(0)}/(1 - 2K_{xc}A^{(0)}) \ . \tag{56d}$$

The result equation (56b) is a consequence of time-reversal symmetry.[14]

For the static case, $\omega = 0$, $\chi_{33}^{P}(q,0)$ was derived by von Barth and Hedin[15] and $\epsilon^{P}(q,0)$ by Hedin and Lundquist,[4] with $K_{xc}, G_{xc\parallel}^{(2)}$ in the general local form as above. For $\omega \neq 0$, $\epsilon^{P}(q,\omega)$ with K_{xc} as the paramagnetic limit of (43b) was derived by Singhal and Callaway.[16]

From $\epsilon(q,\omega)$ given by Eq. (54) we can compute the plasmon dispersion relation

$$\omega_{\rm pl}^2(q) = \omega_{\rm pl}^2[1 + A(\zeta)(q/k_F)^2] \ , \tag{57}$$

where $\omega_{\rm pl}^2 = 4\pi ne^2/m$. Using the appropriate limits of $A_{\sigma\sigma'}^{(0)}(q,\omega)$ given in Ref. 14, we obtain

$$A_{\rm LD}(\zeta) = \tfrac{9}{40}(\pi/\alpha r_s)[(1+\zeta)^{5/3} + (1-\zeta)^{5/3}] - \tfrac{1}{8}[(1+\zeta)^{4/3} + (1-\zeta)^{4/3}] \ . \tag{58}$$

Similarly the static limit of $\epsilon(q\omega)$ gives us the screening ξ^2

$$\lim_{q\to 0} q^2\epsilon(q,\omega=0) = \xi^2 k_F^2 \ , \tag{59}$$

and in the present theory we obtain

$$\xi_{\rm LD}^2(\zeta) = \tfrac{1}{2} q_{\rm TF}^2 \left[\frac{(1+\zeta)^{2/3}}{(1+\zeta)^{1/3} - \alpha r_s/\pi} + \frac{(1-\zeta)^{2/3}}{(1-\zeta)^{1/3} - \alpha r_s/\pi} \right] \ , \tag{60}$$

where $q_{\rm TF}^2$ is the usual Thomas-Fermi value $(4\alpha r_s/\pi)$.

The state long-wavelength limit of $\chi_{33}(q\omega)$ is found to be

$$\chi_{33\,\rm LD}(0,0) = 2\chi^{\rm Pauli}(1-\zeta^2)^{2/3} \times \{(1-\zeta^2)^{1/3}[(1+\zeta)^{1/3} + (1-\zeta)^{1/3}] - (\alpha r_s/\pi)[(1+\zeta)^{2/3} + (1-\zeta)^{2/3}]\}^{-1} \ , \tag{61}$$

where $\chi^{\rm Pauli}$ is the usual Pauli susceptibility of the noninteracting electron gas.

B. Homogeneous electron gas in RPA ($T = 0\,^\circ$K)

The above results can be compared with the corresponding results obtained for the electron gas in Ref. 14, where the relevant vertex functions analogous to Eq. (24) of Sec. II, were set up in the RPA. The local-density model expressed by Eq. (4) should then correspond with these results as far as the ground-state properties are concerned. We shall not repeat these derivations here but quote only the final results. We should remark that the vertex equations were solved in Ref. 14 by a variational method; in the limits concerning us here, it was shown by the author[19] that this procedure is exact.

A detailed comparison of the vertex equations of RPA and those obtained here may be in order. The vertex function given by Eq. (24) is very different in its structure compared to the vertex functions in RPA of Refs. 12 and 14. In the uniform electron gas, for

instance, Γ_{RPA} depends on two four-momentum indices k,q, whereas Γ_{SDF} depends on q only. But, for the calculation of the static long-wavelength limit of the response functions for the electron gas, one requires only Γ_{RPA} $(k=k_F,0;q=0)$. In Ref. 19, the integral equation obeyed by Γ_{RPA} is solved exactly in this particular limit. Using this, we obtained the corresponding response functions for the electron gas $\chi_{RPA}(0,0)$. It is found that $\Gamma_{RPA}(k=k_F,0;q=0) \equiv \Gamma^x_{SDF}(q=0)$, where the superscript x indicates that we have employed the local-exchange approximation, Eq. (43). Indeed, we find that $\xi^2_{LD}(\zeta)=\xi^2_{RPA}(\zeta)$ and $\chi_{33\,LD}(0,0)=\chi_{33\,RPA}(0,0)$ for all ζ. These are ground-state properties of the homogeneous system, and as such we expect the correspondence. The plasmon dispersion, Eq. (57), in RPA is found to differ and we find

$$A_{LD}(\zeta)=A_{RPA}(\zeta)-\tfrac{1}{20}[(1+\zeta)^{4/3}+(1-\zeta)^{4/3}] \quad . \tag{62}$$

The spin-wave dispersion relation was calculated by the author[13] and the result is

$$D_{RPA}(\zeta)=\frac{k_F^2}{2m\zeta}\left[1-\frac{9\pi}{25\alpha r_s}\times\left(\frac{(1+\zeta)^{5/3}-(1-\zeta)^{5/3}}{2\zeta}\right)^2\right] \quad . \tag{63}$$

For $\zeta=1$, we have

$$D_{RPA}(\zeta=1)=\frac{k_F^2}{2m}\left[1-2^{1/3}\frac{18\pi}{25\alpha r_s}\right] \quad , \tag{64}$$

which compares very well with the result obtained by Herring.[10] Callaway and Wang[15] calculated D_{LD} for $\zeta=1$ and it is the same as the one obtained in Eq. (49):

$$D_{LD}(\zeta=1)=\frac{k_F^2}{2m}\left[1-2^{1/3}\frac{2\pi}{5\alpha r_s}\right] \quad . \tag{65}$$

Thus,

$$D_{LD}(\zeta=1)=D_{RPA}(\zeta=1)+2^{1/3}\frac{k_F^2}{2m}\frac{8\pi}{25\alpha r} \quad . \tag{66}$$

From (62) and (66), we observe that the dynamical properties even close to the ground state, are not in any definite relationship with the corresponding RPA result, a confirmation of the doubt one may have in the local-density formalism.

IV. SUMMARY AND CONCLUDING REMARKS

The main results of this work are (i) development of a formalism to calculate the time-dependent response functions of an inhomogeneous electron system in the spin-density-functional theory in the same spirit as in the homogeneous system; (ii) this generalizes, in the static limit $(\omega=0)$, the theory of the paramagnetic susceptibility due to Vosko and Perdew; (iii) for $T=0\,^\circ$K, the local-density-functional theory is shown to yield the same answers as the electron gas theory based on the time-dependent Hartree-Fock scheme for the static long-wavelength limit while the dynamical answers are found to be in disagreement; this is similar to the result of Sham and Kohn[3] who studied the density of states at the Fermi surface; (iv) the local-spin-density functional $E_{xc}[n,|\vec{s}|]$ for a slightly inhomogeneous electron system, takes a neat form. Here

$$n(r)=n_0+\tilde{n}(r), \quad \vec{s}(r)=\vec{s}_0+\tilde{\vec{s}}(r) \quad ,$$

with

$$\int \tilde{n}(r)\,d^3r=0=\int \tilde{\vec{s}}(r)\,d^3r \quad ; \tag{67}$$

and, from Eqs. (35) and (36) we obtain

$$E_{xc}[n,|\vec{s}|]=E_{xc}^{(0)}+\frac{1}{2}\int\int K_{xc}(\vec{r}-\vec{r}')\tilde{n}(\vec{r})\tilde{n}(\vec{r}')\,d^3r\,d^3r'+\frac{1}{2}\int\int G_{xc}^{(1)}(\vec{r}-\vec{r}')[\tilde{n}(\vec{r})\tilde{s}_z(\vec{r}')+\tilde{n}(\vec{r}')\tilde{s}_z(\vec{r})]\,d^3r\,d^3r'$$
$$+\frac{1}{2}\int\int G_{xc\parallel}^{(2)}(\vec{r}-\vec{r}')\tilde{s}_z(\vec{r})\tilde{s}_z(\vec{r}')\,d^3r\,d^3r'+\frac{1}{2}\int G_{xc\perp}^{(2)}(\vec{r})[\tilde{s}_x^2(\vec{r})+\tilde{s}_y^2(\vec{r})]\,d^3r \quad . \tag{68}$$

The linear terms drop out in view of (67) and since V_{xc} and W_{xc} in the homogeneous gas are just constants. The spatial dependences shown in (68) for the various coefficients are a consequence of the fact that they pertain to a homogeneous system. The last three terms are of special significance, in that for a slightly inhomogeneous system, even when the homogeneous counterpart is magnetized along the z direction, the inhomogeneity introduces local transverse coupling between the spin directions, as well as a coupling between the density fluctuations and the longitudinal spin-density fluctuations. The transverse spin fluctuations give rise to spin waves in the system.

The work presented here may be considered as the first step towards a time-dependent density-functional theory. The next step, in view of the difficulties faced in this simple theory, is to build into the formalism, contributions to the self energy, $\Sigma(11)$, from the quasiparticles close to the equilibrium state. For one-electron properties, some suggestions in this

direction have already been made by Sham and Kohn[3] and Hedin and Lundquist.[4] A corresponding scheme based on this can be developed using the formalism given in this paper, but we have not yet succeeded in making it elegant enough to yield analytical results, at least in the long-wavelength limit, so as to eliminate the discrepancy found in the present theory. We hope to return to this important question in the near future.

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