Dynamic correlations in electron liquids. I. General formalism

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Time-dependent multiparticle correlation functions and response functions are formulated with the functional derivatives in the grand-canonical ensemble. Equations for the single-particle Green's functions are the Dyson equations describing scattering of the single particles by density-fluctuation excitations. Equations for the density-fluctuation excitations are truncated with a dynamic version of the convolution approximation to triple correlations. A closed set of equations for the Green's functions and excitations are thereby obtained, which constitutes a dynamic hypernetted-chain scheme for the degenerate electron liquids. The formalism is extended also to the description of the spin-dependent response and correlations.

I. INTRODUCTION

In simple metals such as Na and Al, effects of the crystal potentials are weak,[1,2] so that the electron-liquid model[3] is applicable for the valence electrons. The Coulomb-coupling constant,[1,3] $r_s=(3/4\pi n)^{1/3}me^2/\hbar^2$, for such an electron liquid takes on a magnitude greater than unity (typically, $2\leq r_s\leq 6$); it may thus be looked upon as a strongly coupled system.[4] Here, m and e denote the electronic mass and charge; n refers to the number density of electrons.

In a strongly coupled electron liquid, exchange and Coulomb-induced many-body effects exert essential influence on the quasiparticle properties and spectral functions of dynamic excitations. Recent progress in experimental techniques such as strong synchrotron-radiation sources has made it possible to measure various excitation spectra with an improved accuracy: Single-particle excitation spectra have been investigated through angle-resolved photoemission experiments,[5,6] resulting in observation of conduction-band widths much narrower than those predicted in a perturbation-theoretical calculation.[7] The dynamic structure factor $S(\mathbf{k},\omega)$ has been measured in x-ray inelastic scattering experiments[8] with fine resolutions. The double-peak structures observed in the intermediate wave-number regime were attributed primarily to the lattice-structure effects, rather than to universal properties of a strongly correlated electron liquid; a possibility was also pointed out[8] that some of the fine structures observed, e.g., in polycrystalline samples might be correlation induced.

Most of the theories hitherto proposed on the static and dynamic correlations[1,4] are either perturbation theoretical or dependent on intuitive model calculations. Since the coupling constant is larger than unity for metallic electrons, a perturbative method usually fails to describe the correlational properties correctly; one resorts to a self-consistent theory whereby the many-body effects are taken into account in an iterative manner. Such a theory has been developed successfully for a description of static correlations and for the ground-state properties.[4] It is the purpose of the present paper to develop a self-consistent microscopic theory of *dynamic* correlations in strongly coupled, degenerate electron liquids.

We thus analyze the dynamic evolutions of Green's functions in the grand canonical formalism in the presence of an external perturbation. In the analyses, we make an extensive use of the functional derivative technique, which has been a powerful tool in the classical[9] and quantum[10] theories of liquids. The multiparticle response functions are formulated as the functional derivatives of the single-particle Green's function with respect to the external perturbation. We then define and introduce the multiparticle *correlation potentials* as functional derivatives of the self-energy, the interaction part of the quasiparticle energy, with respect to Green's functions; this is done in a way analogous to the correlation potentials in classical liquids, which are defined as the functional derivatives of the correlation part of the free energy with respect to density fields.[9,11,12]

The response functions are expressible in terms of the Green's functions and correlation potentials, and thus form a chain of equations. We may truncate the hierarchy of the multiparticle response and correlations if an approximation is found which enables us to set an independent expression for the correlation potentials in terms of the Green's functions and response functions. We thereby derive a representation of Dyson equations for the single-particle Green's functions; a *dynamic convolution approximation* is introduced for truncation of the three-body correlation potentials. We thus obtain a set of mutually coupled self-consistent equations for the single-particle Green's functions and the density fluctuation excitations. Solutions to those equations for relevant cases of simple metals will be the subjects of subsequent papers.[13,14]

Organization of the paper is as follows: In Sec. II, the self-energy $\Sigma(\mathbf{k},\omega)$ and the dynamic local-field correction[4] (LFC) $G(\mathbf{k},\omega)$ are formulated in terms of the two- and three-body response functions. In Sec. III, those multiparticle response functions are analyzed through a functional-derivative method; approximations are introduced for truncation. Physical implications of the resulting equations are elucidated in Sec. IV, through investigations of their classical ($\hbar\rightarrow 0$) and weak-coupling ($r_s\rightarrow 0$) limits. In Sec. V, the formalism is extended to the spin-dependent response and correlations. Concluding remarks are given in Sec. VI. Some of the calculational details are explained in Appendixes.

II. FORMULATION OF THE PROBLEM

We consider a system of electrons immersed in a uniform neutralizing background of positive charges. The grand-canonical Hamiltonian is expressed as

$$H=\sum_{\sigma}\int d^3r\, \psi_\sigma^\dagger(\mathbf{r})\left[-\frac{\hbar^2}{2m}\nabla^2-\mu\right]\psi_\sigma(\mathbf{r}) +\frac{1}{2}\sum_{\sigma,\sigma'}\int d^3r\int d^3r'\, \psi_\sigma^\dagger(\mathbf{r})\psi_{\sigma'}^\dagger(\mathbf{r}')v(|\mathbf{r}-\mathbf{r}'|) \times\psi_{\sigma'}(\mathbf{r}')\psi_\sigma(\mathbf{r}) , \quad (1)$$

where μ is the chemical potential, $v(r)=e^2/r$ is the Coulomb potential, and $\psi_\sigma^\dagger(\mathbf{r})$ and $\psi_\sigma(\mathbf{r})$ are the creation and annihilation operators for an electron with spin σ. The single-particle Green's function $\mathcal{G}(1,1')$ is defined as

$$\mathcal{G}(1,1')=-(i/2)\sum_{\sigma}\langle T[\psi_\sigma(1)\psi_\sigma^\dagger(1')]\rangle , \quad (2)$$

where T represents the time-ordering operator from right to left, and we use a shorthand collective-index notation $1\equiv(\mathbf{r}_1,t_1)$. We likewise define an average of an arbitrary operator $\mathcal{O}$ in the presence of a nonlocal external potential $\phi(1,1')$ in the ground state $|0\rangle$ as

$$\langle\mathcal{O}(t)\rangle=\langle 0|T[\mathcal{O}_H(t)\mathcal{S}]|0\rangle/\langle 0|\mathcal{S}|0\rangle , \quad (3)$$

where

$$\mathcal{O}_H(t)=e^{iHt/\hbar}\mathcal{O}e^{-iHt/\hbar} , \quad (4)$$

and $\mathcal{S}$ is the scattering matrix given by

$$\mathcal{S}=T\exp\left[-\frac{i}{\hbar}\sum_{\sigma}\int d1\int d1'\,\psi_{H\sigma}^\dagger(1)\phi(1,1')\psi_{H\sigma}(1')\right] . \quad (5)$$

The ν-body response functions $\chi^{(\nu)}(1,1';\ldots;\nu,\nu')$ are then calculated as the functional derivatives of the Green's function with respect to ϕ:

$$\chi^{(\nu)}(1,1';\ldots;\nu,\nu')=\frac{\delta^{\nu-1}}{\delta\phi(\nu,\nu')\cdots\delta\phi(2,2')} \times\sum_{\sigma}\langle T[\psi_\sigma^\dagger(1)\psi_\sigma(1')]\rangle|_{\phi\to 0} . \quad (6)$$

In particular, the density response function is given by $\chi(1,2)=\chi^{(2)}(1^+,1;2^+,2)$, where $1^+=(\mathbf{r}_1,t_1+0)$ and 0 means a positive infinitesimal.

In the absence of the external field, the Green's function obeys the usual equation of motion with the Hamiltonian (1); its Fourier transform $\mathcal{G}(\mathbf{k},\omega)$ is expressed as

$$\mathcal{G}(\mathbf{k},\omega)=[\omega+\mu/\hbar-\hbar k^2/2m-\Sigma(\mathbf{k},\omega)]^{-1} . \quad (7)$$

Here the self-energy $\Sigma(\mathbf{k},\omega)$ is formulated exactly as

$$\Sigma(\mathbf{k},\omega)=-\frac{1}{2}\int\int\frac{d^3q\,dx}{(2\pi)^4}v(q)e^{ix0} \times\chi_{\mathbf{k}+\mathbf{q}/2,\omega+x/2}(\mathbf{q},x)\mathcal{G}^{-1}(\mathbf{k},\omega) , \quad (8)$$

with $v(q)=4\pi e^2/q^2$, and the electron-hole pair response function $\chi_{\mathbf{p},x}(\mathbf{k},\omega)$ is related to the two-body response function as

$$\chi_{\mathbf{p},x}(\mathbf{k},\omega)=\int d(1-1')\int d[(\mathbf{r}_1+\mathbf{r}_1')/2-\mathbf{r}_2]\int d[(t_1+t_1')/2-t_2]\chi^{(2)}(1',1;2^+,2) \times\exp\{-i\mathbf{p}\cdot(\mathbf{r}_1-\mathbf{r}_1')+ix(t_1-t_1') -i\mathbf{k}\cdot[(\mathbf{r}_1+\mathbf{r}_1')/2-\mathbf{r}_2]+i\omega[(t_1+t_1')/2-t_2]\} . \quad (9)$$

The dielectric response function[3,4] $\epsilon(\mathbf{k},\omega)$ is related with the Fourier transform $\chi(\mathbf{k},\omega)$ of the density response function as

$$\epsilon^{-1}(\mathbf{k},\omega)=1+v(k)\chi(\mathbf{k},\omega) =1+v(k)\int\int\frac{d^3p\,dx}{(2\pi)^4}e^{ix0}\chi_{\mathbf{p},x}(\mathbf{k},\omega) . \quad (10)$$

The dynamic LFC, $G(\mathbf{k},\omega)$, is then defined and introduced via[4]

$$\chi(\mathbf{k},\omega)=\chi_L(\mathbf{k},\omega)/\{1-v(k)[1-G(\mathbf{k},\omega)]\chi_L(\mathbf{k},\omega)\} , \quad (11)$$

where the Lindhard polarizability[15] is given by

$$\chi_L(\mathbf{k},\omega)=-\frac{1}{\hbar}\int\frac{d^3p}{(2\pi)^3}\frac{\Delta_{\mathbf{p}}^{\mathbf{k}}F_{\mathbf{p}}}{\omega-\hbar\mathbf{k}\cdot\mathbf{p}/m+i0\,\mathrm{sgn}(\omega)} , \quad (12)$$

and $F_{\mathbf{p}}$ is the momentum distribution function . In Eq. (12), $\Delta_{\mathbf{p}}^{\mathbf{k}}$ is a difference operator acting on a $\mathbf{p}$-dependent function $f(\mathbf{p})$ as $\Delta_{\mathbf{p}}^{\mathbf{k}}f(\mathbf{p})=f(\mathbf{p}+\mathbf{k}/2)-f(\mathbf{p}-\mathbf{k}/2)$. The LFC is expressed in terms of the three-body density response function as

$$G(\mathbf{k},\omega)=-\frac{i\hbar}{n\chi(\mathbf{k},\omega)}\int\int\frac{d^3q\,dx}{(2\pi)^4}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}e^{ix0} \times\chi^{(3)}(\mathbf{k}-\mathbf{q},\omega-x;\mathbf{q},x) , \quad (13)$$

where

$$\chi^{(3)}(\mathbf{k},\omega;\mathbf{q},x) =\int d(1-2)\int d(3-2)\chi^{(3)}(1^+,1;2^+,2;3^+,3) \times\exp[-i\mathbf{k}\cdot(\mathbf{r}_1-\mathbf{r}_2)+i\omega(t_1-t_2) -i\mathbf{q}\cdot(\mathbf{r}_3-\mathbf{r}_2)+ix(t_3-t_2)] . \quad (14)$$

A derivation of Eq. (13) is described in Appendix A; a proof is given also that Eq. (13) is exact in the high-frequency or long-wavelength limit.

The functions $\mathcal{G}(\mathbf{k},\omega)$ and $\chi(\mathbf{k},\omega)$ retain many-body effects in $\Sigma(\mathbf{k},\omega)$ and $G(\mathbf{k},\omega)$, which in turn depend on two- and three-body response functions. If the latter functions are expressed in terms of $\mathcal{G}(\mathbf{k},\omega)$ and $\chi(\mathbf{k},\omega)$, truncation is completed and we obtain a closed set of equations; this will be done in the next section.

III. CALCULATIONS OF $\Sigma(\mathbf{k},\omega)$ AND $G(\mathbf{k},\omega)$

For analyses of the multiparticle response functions, we find it useful to introduce ν-body correlation potentials $\Xi^{(\nu)}(1,1';\ldots;\nu,\nu')$, defined as the functional derivatives of Σ with respect to $\mathcal{G}$,

$$\Xi^{(\nu)}(1,1';\ldots;\nu,\nu')=\frac{\delta^{\nu-1}}{\delta\mathcal{G}(\nu,\nu')\cdots\delta\mathcal{G}(2,2')}\Sigma(1,1') \ . \quad (15)$$

Physically, $\Xi^{(\nu)}$ represent the effective ν-body interactions stemming from interparticle correlations. In particular, $\Xi^{(2)}$ is the same as what has been called the irreducible particle-hole interaction,[16] except for the direct Coulomb interaction contained in the latter.

We begin with an exact equation for the electron-hole pair response function $\chi^{(2)}(1',1;2^+,2)$, derived in Appendix B:

$$\chi^{(2)}(1',1;2^+,2)=-2i\hbar^{-1}\mathcal{G}(1,2)\mathcal{G}(2,1')-2i\hbar^{-1}\mathcal{G}(1,\bar{3})\mathcal{G}(\bar{3},1')v(\bar{3},\bar{4})\chi(\bar{4},2)$$
$$+\mathcal{G}(1,\bar{3})\mathcal{G}(\bar{3}',1')\Xi^{(2)}(\bar{3},\bar{3}';\bar{4},\bar{4}')\chi^{(2)}(\bar{4}',\bar{4};2^+,2) \ . \quad (16)$$

Here, $v(3,4)=v(|\mathbf{r}_3-\mathbf{r}_4|)\delta(t_3-t_4)$, and integrations with respect to the barred indices are implied. Equation (16) is diagrammatically represented in Fig. 1, where thick lines refer to $\chi^{(2)}$, a directed (thin) line $\mathcal{G}$, a double line χ, a dashed line v, and a shaded square $\Xi^{(2)}$; a solid circle implies an integration over the index.

We now introduce a local approximation, such that

$$\Xi^{(2)}(3,3';4,4')=\tilde{\Xi}^{(2)}(3,4)\delta(3,3')\delta(4,4') \quad (17)$$

in Eq. (16); $\delta(3,3')$ refers to a four-dimensional δ function. This approximation may be justified through the following arguments: Since $\Xi^{(2)}(3,3';4,4')$ is short ranged[17] in the sense that $|\mathbf{r}_3-\mathbf{r}_3'|$ and $|\mathbf{r}_4-\mathbf{r}_4'|$ have spatial extents on the order of the Fermi distance, $k_F^{-1}=(3\pi^2 n)^{-1/3}$, it may be regarded as spatially local in comparison with the long-range quality of the Coulomb interaction $v(3,4)$ in Eq. (16). Analogously, the temporal scales of variations in $\Xi^{(2)}$ may be confined within the Fermi time $\sim\hbar/E_F$, which we assume to be smaller significantly than the characteristic times of the long-lived density-fluctuation excitations contained in $\chi^{(2)}$; $E_F=(\hbar k_F)^2/2m$ is the Fermi energy.

In the local approximation (17), Eq. (16) can be solved for $\chi^{(2)}(1',1;2^+,2)$ in the form

$$\chi_{\mathbf{p},x}(\mathbf{k},\omega)=-2i\hbar^{-1}\mathcal{G}(\mathbf{p}-\mathbf{k}/2,x-\omega/2)$$
$$\times\mathcal{G}(\mathbf{p}+\mathbf{k}/2,x+\omega/2)/\tilde{\epsilon}(\mathbf{k},\omega), \quad (18)$$

where

$$\tilde{\epsilon}^{-1}(\mathbf{k},\omega)=\chi(\mathbf{k},\omega)/\chi_0(\mathbf{k},\omega), \quad (19)$$

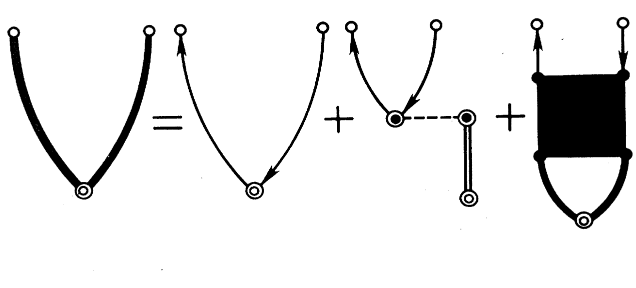

FIG. 1. Diagrammatic representation of Eq. (16).

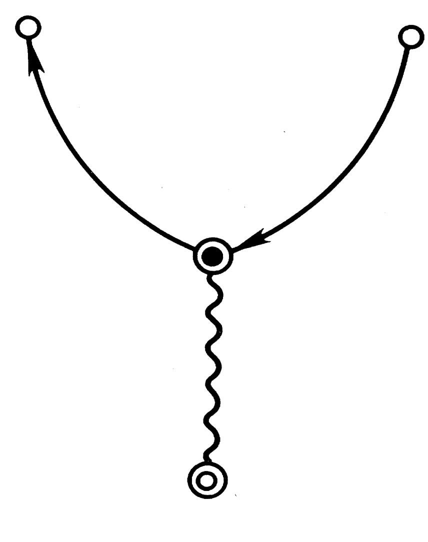

FIG. 2. Diagrammatic representation of Eq. (18).

$$\chi_0(\mathbf{k},\omega) = -\frac{2i}{\hbar}\int\int \frac{d^3p\,dx}{(2\pi)^4}\mathcal{G}(\mathbf{p}-\mathbf{k}/2, x-\omega/2) \times \mathcal{G}(\mathbf{p}+\mathbf{k}/2, x+\omega/2) \; . \tag{20}$$

After the local approximation, Eq. (16) reduces to Eq. (18) depicted in Fig. 2, where a wavy line represents $\tilde{\epsilon}^{-1}$. Substitution of Eq. (18) in Eq. (8) yields

$$\Sigma(\mathbf{k},\omega) = \frac{i}{\hbar}\int\int \frac{d^3q\,dx}{(2\pi)^4} e^{ix0}\frac{v(q)}{\tilde{\epsilon}(\mathbf{q},x)}\mathcal{G}(\mathbf{k}+\mathbf{q},\omega+x) \; . \tag{21}$$

The three-body density response function is calculated as (see Appendix B)

$$\chi^{(3)}(1^+,1;2^+,2;3^+,3) = -(\hbar/4)\chi^{(2)}(1^+,1;\bar{4},\bar{4}')\chi^{(2)}(\bar{5}',\bar{5};2^+,2)\chi^{(2)}(\bar{6}',\bar{6};3^+,3) \times[\mathcal{G}^{-1}(\bar{4},\bar{6})\mathcal{G}^{-1}(\bar{6}',\bar{5})\mathcal{G}^{-1}(\bar{5}',\bar{4}') + \mathcal{G}^{-1}(\bar{4},\bar{5})\mathcal{G}^{-1}(\bar{5}',\bar{6})\mathcal{G}^{-1}(\bar{6}',\bar{4}') + \Xi^{(3)}(\bar{4},\bar{4}';\bar{5},\bar{5}';\bar{6},\bar{6}')] \; . \tag{22}$$

Here again, we assume the local approximations, Eq. (17) and $\Xi^{(3)}(1,1';2,2';3,3') = \tilde{\Xi}^{(3)}(1,2,3)\delta(1,1')\delta(2,2')\delta(3,3')$, to the correlation potentials; then Eq. (22) becomes

$$\chi^{(3)}(1^+,1;2^+,2;3^+,3) = \chi_0^{(3)}(\bar{4},\bar{5},\bar{6})\tilde{\epsilon}^{-1}(1,\bar{4}) \times\tilde{\epsilon}^{-1}(\bar{5},2)\tilde{\epsilon}^{-1}(\bar{6},3) - (\hbar/4)\tilde{\Xi}^{(3)}(\bar{4},\bar{5},\bar{6}) \times\chi(1,\bar{4})\chi(\bar{5},2)\chi(\bar{6},3) \; , \tag{23}$$

where

$$\chi_0^{(3)}(1,2,3) = -2i\hbar^{-2}[\mathcal{G}(1,3)\mathcal{G}(3,2)\mathcal{G}(2,1) + \mathcal{G}(1,2)\mathcal{G}(2,3)\mathcal{G}(3,1)] \; . \tag{24}$$

Figure 3 depicts Eq. (23), where a shaded triangle represents $\tilde{\Xi}^{(3)}$.

The three-body correlation potential $\tilde{\Xi}^{(3)}(4,5,6)$ in the last term of Eq. (23) depends intrinsically on the triple and higher-order correlation functions. If we ignore this term in Eq. (23) and substitute the remainder in Eq. (13), we obtain

$$G(\mathbf{k},\omega) = -\frac{i\hbar}{n\chi_0(\mathbf{k},\omega)}\int\int\frac{d^3q\,dx}{(2\pi)^4}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}e^{ixo} \times\chi_0^{(3)}(\mathbf{k}-\mathbf{q},\omega-x;\mathbf{q},x) \times\tilde{\epsilon}^{-1}(\mathbf{k}-\mathbf{q},\omega-x) \times\tilde{\epsilon}^{-1}(\mathbf{q},x) \; , \tag{25}$$

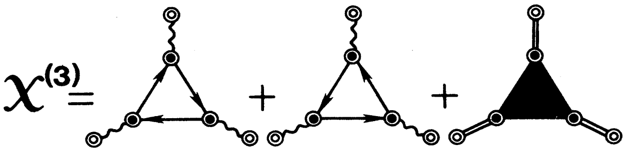

FIG. 3. Diagrammatic representation of Eq. (23).

where

$$\chi_0^{(3)}(\mathbf{k},\omega;\mathbf{q},x) = -\frac{2i}{\hbar^2}\int\int\frac{d^3p\,dy}{(2\pi)^4}[\mathcal{G}(\mathbf{p},y)\mathcal{G}(\mathbf{p}+\mathbf{k},y+\omega) \times\mathcal{G}(\mathbf{p}-\mathbf{q},y-x) + \{(\mathbf{k},\omega)\leftrightarrow(\mathbf{q},x)\}] \tag{26}$$

is a Fourier transform of $\chi_0^{(3)}(1,2,3)$ and hence depends only on the single-particle Green's functions. Hierarchies of the many-body correlations and response functions are thus truncated at the second stage involving density-fluctuation excitations. Consequently, Eqs. (7), (11), (21), and (25) constitute a closed set of equations for $\mathcal{G}(\mathbf{k},\omega)$ and $\chi(\mathbf{k},\omega)$.

As we shall show in the subsequent section, the neglect of the last term in Eq. (23) amounts to adopting a dynamic version of the convolution approximation to the three-body density response. Significance of the convolution approximation for the long-ranged Coulombic system and its relation with the hypernetted-chain approximation have been well elucidated;[11,12] it ensures perfect screening and thereby maintains the short ranged quality of the correlation potentials. We may thus call Eq. (25) the LFC in the *dynamic hypernetted-chain approximation.* Inclusion of nonvanishing contributions from $\tilde{\Xi}^{(3)}(4,5,6)\neq 0$ would then lead to a modification and improvement on such an approximation.

IV. PHYSICAL PROPERTIES OF THE SELF-CONSISTENT EQUATIONS

In this section, we intend to elucidate some of the physical contents in the self-consistent equations derived in the preceding section. We shall do so through investigation of those equations in various limits.

Let us first take up the classical limit ($\hbar\to 0$) of Eqs. (7) and (21) for the single-particle Green's function. Following the notation in Kadanoff and Baym,[10] we decompose each of $\mathcal{G}(1,1')$ and $\Sigma(1,1')$ into two analytic functions, $\mathcal{G}^{\gtrless}(1,1')$ and $\Sigma^{\gtrless}(1,1')$, in accord with $t_1 \gtrless t_1'$, and rewrite these functions in terms of new variables, $\mathbf{r}=\mathbf{r}_1-\mathbf{r}_1'$, $t=t_1-t_1'$, $\mathbf{R}=(\mathbf{r}_1+\mathbf{r}_1')/2$, and $T=(t_1+t_1')/2$; we then define their Fourier transforms via, e.g.,

$$\mathcal{G}^{\gtrless}(\mathbf{p},\omega;\mathbf{R},T)=\pm i\int d^3r\int dt\,\exp(-i\mathbf{p}\cdot\mathbf{r}/\hbar+i\omega t)\times\mathcal{G}^{\gtrless}(\mathbf{r},t;\mathbf{R},T)\ , \tag{27}$$

and the distribution function as

$$f(\mathbf{p};\mathbf{R},T)=\int\frac{d\omega}{2\pi}\mathcal{G}^{<}(\mathbf{p},\omega;\mathbf{R},T)\ . \tag{28}$$

The equation of motion for $f(\mathbf{p};\mathbf{R},T)$ is derived as[10]

$$\left[\frac{\partial}{\partial T}+\frac{\mathbf{p}}{m}\cdot\frac{\partial}{\partial\mathbf{R}}-\left[\frac{\partial}{\partial\mathbf{R}}\phi_H(\mathbf{R},T)\right]\cdot\frac{\partial}{\partial\mathbf{p}}\right]f(\mathbf{p};\mathbf{R},T)=\int\frac{d\omega}{2\pi}[\mathcal{G}^{>}(\mathbf{p},\omega;\mathbf{R},T)\Sigma^{<}(\mathbf{p},\omega;\mathbf{R},T)-\mathcal{G}^{<}(\mathbf{p},\omega;\mathbf{R},T)\Sigma^{>}(\mathbf{p},\omega;\mathbf{R},T)]\ , \tag{29}$$

where we have assumed a slowly varying external field $\phi(1,1')=\phi(1)\delta(1,1')$; $\phi_H(1)=\phi(1)+v(1,\bar{2})\times\sum_\sigma\langle\psi_\sigma^\dagger(\bar{2})\psi_\sigma(\bar{2})\rangle$ is its Hartree field. Substitution of Eq. (21) in the right-hand side of Eq. (29) yields

$$\hbar^{-2}\int\int\frac{d^3q\,dx}{(2\pi)^2}v^2(q)\tilde{A}(\mathbf{p}+\hbar\mathbf{q}/2;\mathbf{q},x)\times[\tilde{S}(\mathbf{q},x)f(\mathbf{p}+\hbar\mathbf{q};\mathbf{R},T)-\tilde{S}(\mathbf{q},-x)f(\mathbf{p};\mathbf{R},T)]\ . \tag{30}$$

Here we have assumed separation between fast and slow variations so that

$$\int dx\ \mathcal{G}^{\gtrless}(\mathbf{p}+\hbar\mathbf{k}/2,x+\omega/2;\mathbf{R},T)\mathcal{G}^{\lessgtr}(\mathbf{p}-\hbar\mathbf{k}/2,x-\omega/2;\mathbf{R},T)=[1-f(\mathbf{p}\pm\hbar\mathbf{k}/2;\mathbf{R},T)]f(\mathbf{p}\mp\hbar\mathbf{k}/2;\mathbf{R},T)\tilde{A}(\mathbf{p};\mathbf{k},\omega)\ , \tag{31}$$

where

$$\tilde{A}(\mathbf{p};\mathbf{k},\omega)=\int dx\ A(\mathbf{p}+\hbar\mathbf{k}/2,x+\omega/2)A(\mathbf{p}-\hbar\mathbf{k}/2,x-\omega/2)\ , \tag{32}$$

and

$$A(\mathbf{p},\omega)=\tfrac{1}{2}\sum_\sigma\int d(1-1')\exp[-i\mathbf{p}\cdot(\mathbf{r}_1-\mathbf{r}_1')/\hbar+i\omega(t_1-t_1')]\langle[\psi_\sigma(1)\psi_\sigma^\dagger(1')+\psi_\sigma^\dagger(1')\psi_\sigma(1)]\rangle \tag{33}$$

is the spectral function for the single-particle Green's function. In Eq. (30), we have introduced $\tilde{S}(\mathbf{q},x)$ through the relation

$$\tilde{S}(\mathbf{q},x)-\tilde{S}(\mathbf{q},-x)=-[\hbar/\pi v(q)]\mathrm{Im}\tilde{\epsilon}_R^{-1}(\mathbf{q},x)\ , \tag{34}$$

where $\tilde{\epsilon}_R(\mathbf{q},x)$ is the retarded counterpart to $\tilde{\epsilon}(\mathbf{q},x)$. In deriving Eq. (30), we have also assumed $f(\mathbf{p};\mathbf{R},T)\ll 1$, valid in the limit $\hbar\to 0$.

Expanding Eq. (30) in powers of $\hbar$ and keeping only the leading terms, we arrive at the Fokker-Planck equation,[18]

$$\left[\frac{\partial}{\partial T}+\frac{\mathbf{p}}{m}\cdot\frac{\partial}{\partial\mathbf{R}}\left[\frac{\partial}{\partial\mathbf{R}}\phi_H(\mathbf{R},T)\right]\cdot\frac{\partial}{\partial\mathbf{p}}-\frac{\partial}{\partial\mathbf{p}}\cdot\overleftrightarrow{\mathbf{D}}(\mathbf{p})\cdot\frac{\partial}{\partial\mathbf{p}}-\frac{\partial}{\partial\mathbf{p}}\cdot\mathbf{F}(\mathbf{p})\right]f(\mathbf{p};\mathbf{R},T)=0\ , \tag{35}$$

where the diffusion and friction coefficients, $\overleftrightarrow{\mathbf{D}}(\mathbf{p})$ and $\mathbf{F}(\mathbf{p})$, are given by

$$\overleftrightarrow{\mathbf{D}}(\mathbf{p})=\pi\int\int\frac{d^3q\,dx}{(2\pi)^3}\mathbf{q}\mathbf{q}v^2(q)\tilde{S}(\mathbf{q},x)\tilde{A}(\mathbf{p};\mathbf{q},x)\ , \tag{36}$$

$$\mathbf{F}(\mathbf{p})=-\int\int\frac{d^3q\,dx}{(2\pi)^3}\mathbf{q}v(q)\mathrm{Im}\tilde{\epsilon}_R^{-1}(\mathbf{q},x)\tilde{A}(\mathbf{p};\mathbf{q},x)\ . \tag{37}$$

Hence, Eqs. (7) and (21) are consistent with the classical notion of single particles being scattered by field fluctuations.

Next, we show that the approximation $\tilde{\Xi}^{(3)}=0$ in Eq. (23) at vanishing frequencies, i.e.,

$$\chi^{(3)}(\mathbf{k},0;\mathbf{q},0)=\chi_0^{(3)}(\mathbf{k},0;\mathbf{q},0)\tilde{\epsilon}^{-1}(\mathbf{k},0)\times\tilde{\epsilon}^{-1}(\mathbf{q},0)\tilde{\epsilon}^{-1}(\mathbf{k}+\mathbf{q},0) \tag{38}$$

reduces in the classical limit to the convolution approximation for the three-body correlation function.[11,12] To do so, we work in the grand-canonical ensemble with $\phi(1,1')=\phi(\mathbf{r}_1)\delta(1,1')$. The Ursell functions $U^{(\nu)}(\mathbf{r}_1,\ldots,\mathbf{r}_\nu)$ defined by[12]

$$U^{(\nu)}(\mathbf{r}_1,\ldots,\mathbf{r}_\nu)=(-k_BT)^{\nu-1}\frac{\delta^{\nu-1}}{\delta\phi(\mathbf{r}_\nu)\cdots\delta\phi(\mathbf{r}_2)}\times\sum_\sigma\langle\psi_\sigma^\dagger(\mathbf{r}_1)\psi_\sigma(\mathbf{r}_1)\rangle|_{\phi\to 0} \tag{39}$$

have Fourier transforms

$$\tilde{U}^{(2)}(\mathbf{k})=-k_BT\chi(\mathbf{k},0)\ , \tag{40}$$

$$\tilde{U}^{(3)}(\mathbf{k},\mathbf{q})=(k_BT)^2\chi^{(3)}(\mathbf{k},0;\mathbf{q},0)\ . \tag{41}$$

One can likewise prove

$$\lim_{\hbar\to 0}\chi_0(\mathbf{k},0)=-n/k_BT\ , \tag{42}$$

$$\lim_{\hbar\to 0}\chi_0^{(3)}(\mathbf{k},0;\mathbf{q},0)=n/(k_BT)^2\ , \tag{43}$$

so that the classical limit of Eq. (38) turns into a convolution-approximation formula,[11,12]

$$\tilde{U}^{(3)}(\mathbf{k},\mathbf{q})=n^{-2}\tilde{U}^{(2)}(\mathbf{k})\tilde{U}^{(2)}(\mathbf{q})\tilde{U}^{(2)}(\mathbf{k}+\mathbf{q})\ , \tag{44}$$

with the aid of Eq. (19).

Finally, we consider the weak-coupling limit ($r_s\to 0$) of Eq. (25). In this limit, we may set $\tilde{\varepsilon}(\mathbf{k},\omega)=1$ and $\mathcal{G}(\mathbf{k},\omega)=[\omega-\hbar(k^2-k_F^2)/2m+i0\,\mathrm{sgn}(\omega)]^{-1}$. After performing the frequency integrals in Eq. (25), we move over to the retarded boundary conditions by shifting all the poles in the upper half of the complex ω plane into the lower half. The result then is

$$G(\mathbf{k},\omega)=-\frac{2}{n\hbar\chi_L(\mathbf{k},\omega)}\int\int\frac{d^3p\,d^3p'\,\mathbf{k}\cdot(\mathbf{p}-\mathbf{p}')}{(2\pi)^6|\mathbf{p}-\mathbf{p}'|^2}\frac{\Delta_{\mathbf{p}}^{\mathbf{k}}\Theta(k_F-p)\Delta_{\mathbf{p}'}^{\mathbf{k}}\Theta(k_F-p')}{\omega-\hbar\mathbf{k}\cdot\mathbf{p}/m+i0}\ , \tag{45}$$

where $\Theta(x)=[1+\mathrm{sgn}(x)]/2$ is the unit step function. Equation (45) is the same as the LFC derived originally by Toigo and Woodruff[19] through the analysis of lowest-order proper-polarization diagrams. Hence, Eq. (25) correctly describes the exchange effect in the weak-coupling limit.

V. SPIN RESPONSE

Thus far we have been concerned with formulation of the single-particle Green's function and the density response function. The formalism can be extended to describe the spin-dependent properties. To do so, we recover the spin indices and consider the spin-dependent Green's functions,

$$\mathcal{G}_\sigma(1,1')=-i\langle T[\psi_\sigma(1)\psi_\sigma^\dagger(1')]\rangle\ , \tag{46}$$

which in conjunction with a spin-dependent external field $\phi_\sigma(1,1')$ introduced in Eq. (5) lead to the response functions,

$$\chi_{\sigma\tau\ldots\upsilon}^{(\nu)}(1,1';\ldots;\nu,\nu')=\frac{\delta^{\nu-1}}{\delta\phi_\upsilon(\nu,\nu')\cdots\delta\phi_\tau(2,2')}\langle T[\psi_\sigma^\dagger(1)\psi_\sigma(1')]\rangle|_{\phi_\sigma\to 0}\ . \tag{47}$$

In particular, $\chi_{\sigma\tau}(1,2)=\chi_{\sigma\tau}^{(2)}(1^+,1;2^+,2)$ is the density-density response function between the spin components σ and τ, and $\zeta(1,2)=\sum_{\sigma,\tau}(2\delta_{\sigma\tau}-1)\chi_{\sigma\tau}(1,2)$ is the spin response function.

Spin-dependent LFC's are introduced through an extension of Eq. (11), i.e.,

$$\chi_{\sigma\tau}(\mathbf{k},\omega)=\chi_{L\sigma}(\mathbf{k},\omega)\left[\delta_{\sigma\tau}+\sum_{\upsilon}v(k)[1-G_{\sigma\upsilon}(\mathbf{k},\omega)]\chi_{\upsilon\tau}(\mathbf{k},\omega)\right]\ , \tag{48}$$

where $\chi_{L\sigma}(\mathbf{k},\omega)$ is given by Eq. (12) with $F_{\mathbf{p}}$ replaced by $F_{\mathbf{p}\sigma}$, the momentum distribution function of the spin component σ. The LFC's are then formulated as

$$G_{\sigma\tau}(\mathbf{k},\omega)=-\frac{2i\hbar}{n\chi_{0\tau}(\mathbf{k},\omega)}\sum_{\upsilon}\int\int\frac{d^3q\,dx}{(2\pi)^4}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}e^{ix0}\chi_{0\tau}^{(3)}(\mathbf{k}-\mathbf{q},\omega-x;\mathbf{q},x)\tilde{\varepsilon}_{\sigma\tau}^{-1}(\mathbf{k}-\mathbf{q},\omega-x)\tilde{\varepsilon}_{\upsilon\tau}^{-1}(\mathbf{q},x)\ , \tag{49}$$

by following a procedure analogous to that in Appendix A. Here we recall

$$\tilde{\varepsilon}_{\sigma\tau}^{-1}(\mathbf{k},\omega)=\chi_{\sigma\tau}(\mathbf{k},\omega)/\chi_{0\sigma}(\mathbf{k},\omega)\ , \tag{50}$$

with

$$\chi_{0\sigma}(\mathbf{k},\omega)=-\frac{i}{\hbar}\int\int\frac{d^3p\,dx}{(2\pi)^4}\mathcal{G}_\sigma(\mathbf{p}-\mathbf{k}/2,x-\omega/2)\mathcal{G}_\sigma(\mathbf{p}+\mathbf{k}/2,x+\omega/2)\ , \tag{51}$$

$$\chi_{0\sigma}^{(3)}(\mathbf{k},\omega;\mathbf{q},x)=-\frac{i}{\hbar^2}\int\int\frac{d^3p\,dy}{(2\pi)^4}[\mathcal{G}_\sigma(\mathbf{p},y)\mathcal{G}_\sigma(\mathbf{p}+\mathbf{k},y+\omega)\mathcal{G}_\sigma(\mathbf{p}-\mathbf{q},y-x)+\{(\mathbf{k},\omega)\leftrightarrow(\mathbf{q},x)\}]\ . \tag{52}$$

We have assumed the dynamic convolution approximations of Sec. III in the calculation of Eq. (49).

For the electron liquid in the paramagnetic state where $G_{\sigma\sigma}=G_{-\sigma-\sigma}$ and $G_{\sigma-\sigma}=G_{-\sigma\sigma}$, the spin LFC, $J(\mathbf{k},\omega)$, introduced via[12]

$$\zeta(\mathbf{k},\omega)=\chi_L(\mathbf{k},\omega)/[1+v(k)J(\mathbf{k},\omega)\chi_L(\mathbf{k},\omega)]\ , \tag{53}$$

is calculated as

$$J(\mathbf{k},\omega)=-\frac{i\hbar}{n\chi_0(\mathbf{k},\omega)}\int\int\frac{d^3q\,dx}{(2\pi)^4}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}e^{ix0}\times\chi_0^{(3)}(\mathbf{k}-\mathbf{q},\omega-x;\mathbf{q},x)\times\xi^{-1}(\mathbf{k}-\mathbf{q},\omega-x)\tilde{\varepsilon}^{-1}(\mathbf{q},x)\ , \tag{54}$$

where

$$\xi^{-1}(\mathbf{k},\omega)=\zeta(\mathbf{k},\omega)/\chi_0(\mathbf{k},\omega)\ . \tag{55}$$

Those equations for $\mathcal{G}(\mathbf{k},\omega)$ and $\chi(\mathbf{k},\omega)$ remain unchanged. Hence, Eqs. (53) and (54), together with Eqs. (7), (11), (21), and (25), constitute a closed set of equations for $\mathcal{G}(\mathbf{k},\omega)$, $\chi(\mathbf{k},\omega)$, and $\zeta(\mathbf{k},\omega)$.

VI. CONCLUSION

We have derived a set of self-consistent integral equations describing the dynamic behaviors of the single-particle Green's functions $\mathcal{G}_\sigma(\mathbf{k},\omega)$, the charge-density response $\chi(\mathbf{k},\omega)$, and the spin-density response $\zeta(\mathbf{k},\omega)$. The equations for $\mathcal{G}_\sigma(\mathbf{k},\omega)$ are Dyson equations, which reduce in the classical limit to the Fokker-Planck equations describing scatterings of single particles in the field fluctuations. Truncation of the hierarchy has been achieved through introduction of the dynamic convolution approximations in the equations for the response functions. This scheme of truncation may thus correspond to the dynamic hypernetted-chain approximation to the integral equations. Solutions to those equations under specific circumstances will be considered in the subsequent publications.[13,14]

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APPENDIX A: DERIVATION OF EQ. (13)

We define an equal-time electron-hole pair response function $\chi_{\mathrm{p}}(\mathbf{k},\omega)$ as

$$\chi_{\mathrm{p}}(\mathbf{k},\omega)=\int \frac{dx}{2\pi}e^{ix0}\chi_{\mathrm{p},x}(\mathbf{k},\omega)\ , \tag{A1}$$

which obeys the equation of motion:

$$(\omega-\hbar\mathbf{k}\cdot\mathbf{p}/m)\chi_{\mathrm{p}}(\mathbf{k},\omega)=-\Delta_{\mathrm{p}}^{\mathbf{k}}F_{\mathrm{p}}[1+v(k)\chi(\mathbf{k},\omega)]-i\hbar\int\int\frac{d^3q\,dx}{(2\pi)^4}e^{ix0}v(q)\Delta_{\mathrm{p}}^{\mathbf{q}}\chi_{\mathrm{p}}^{(3)}(\mathbf{k}-\mathbf{q},\omega-x;\mathbf{q},x)\ . \tag{A2}$$

Here

$$\begin{aligned}\chi_{\mathrm{p}}^{(3)}(\mathbf{k},\omega;\mathbf{q},x)=&\int d(\mathbf{r}_1-\mathbf{r}_1')\int d[(\mathbf{r}_1+\mathbf{r}_1')/2-\mathbf{r}_2]\\ &\times\int d(t_1-t_2)\int d(3-2)\chi^{(3)}(\mathbf{r}_1',t_1+0;\mathbf{r}_1,t_1;3^+,3;2^+,2)\\ &\times\exp\{-i\mathbf{p}\cdot(\mathbf{r}_1-\mathbf{r}_1')-i\mathbf{k}\cdot[(\mathbf{r}_1+\mathbf{r}_1')/2-\mathbf{r}_2]\\ &+i\omega(t_1-t_2)-i\mathbf{q}\cdot(\mathbf{r}_3-\mathbf{r}_2)+ix(t_3-t_2)\}\ .\end{aligned} \tag{A3}$$

The LFC is introduced via

$$(\omega-\hbar\mathbf{k}\cdot\mathbf{p}/m)\chi_{\mathrm{p}}(\mathbf{k},\omega)=-\Delta_{\mathrm{p}}^{\mathbf{k}}F_{\mathrm{p}}\{1+v(k)[1-G(\mathbf{k},\omega)]\chi(\mathbf{k},\omega)\}\ , \tag{A4}$$

in accord with Eq. (11); it is determined through comparison between Eqs. (A2) and (A4) as follows: We first multiply Eq. (A2) by $\hbar\mathbf{k}\cdot\mathbf{p}/m$, integrate the resulting equation over $\mathbf{p}$, and then obtain

$$(\omega^2-\omega_p^2)\chi(\mathbf{k},\omega)=\int\frac{d^3p}{(2\pi)^3}\left[\frac{\hbar\mathbf{k}\cdot\mathbf{p}}{m}\right]\chi_{\mathrm{p}}(\mathbf{k},\omega)+\frac{nk^2}{m}+\frac{i\hbar\omega_p^2}{n}\int\int\frac{d^3q\,dx}{(2\pi)^4}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}e^{ix0}\chi^{(3)}(\mathbf{k}-\mathbf{q},\omega-x;\mathbf{q},x)\ , \tag{A5}$$

where $\omega_p=(4\pi ne^2/m)^{1/2}$ is the plasma frequency. In deriving Eq. (A5), we have used the continuity equation,

$$\omega\chi(\mathbf{k},\omega)=\int\frac{d^3p}{(2\pi)^3}\frac{\hbar\mathbf{k}\cdot\mathbf{p}}{m}\chi_{\mathrm{p}}(\mathbf{k},\omega)\ , \tag{A6}$$

which can be obtained by integrating Eq. (A2) over $\mathbf{p}$. Applying the same procedure to Eq. (A4) and setting the resulting equation equal to Eq. (A5), we obtain Eq. (13).

We now show that Eq. (13) is exact for either $\omega\to\infty$ or $k\to 0$. Multiplying Eq. (A2) by $[\omega-\hbar\mathbf{k}\cdot\mathbf{p}/m+i0\,\mathrm{sgn}(\omega)]^{-1}$ and integrating it over $\mathbf{p}$, we obtain Eq. (11), with $G(\mathbf{k},\omega)$ given by

$$G(\mathbf{k},\omega)=\frac{i}{v(k)\chi_L(\mathbf{k},\omega)\chi(\mathbf{k},\omega)}\int\int\int\frac{d^3p\,d^3q\,dx}{(2\pi)^7}e^{ix0}\frac{v(q)\Delta_{\mathrm{p}}^{\mathbf{q}}}{\omega-\hbar\mathbf{k}\cdot\mathbf{p}/m+i0\,\mathrm{sgn}(\omega)}\chi_{\mathrm{p}}^{(3)}(\mathbf{k}-\mathbf{q},\omega-x;\mathbf{q},x)\ , \tag{A7}$$

which is thus an exact expression defined in Eq. (11). Substituting the expansion

$$[\omega-\hbar\mathbf{k}\cdot\mathbf{p}/m+i0\,\mathrm{sgn}(\omega)]^{-1}=\omega^{-1}(1+\hbar\mathbf{k}\cdot\mathbf{p}/m\omega+\cdots), \tag{A8}$$

in $\chi_L(\mathbf{k},\omega)$ and in the integrand of Eq. (A7), and keeping only the leading terms in the expansion, we recover Eq. (13). Since Eq. (A8) is correct either in the limit $\omega\rightarrow\infty$ or $k\rightarrow 0$, Eq. (13) becomes exact in these limits. In fact, we can prove that $\chi(\mathbf{k},\omega)$ satisfies the third-frequency moment sum rule[4] by virtue of Eq. (13), through the same method as that elaborated in Niklasson.[20]

APPENDIX B: DERIVATION OF EQS. (16) AND (22)

The inverse $\mathcal{G}^{-1}$ of the single-particle Green's function is defined via

$$\mathcal{G}^{-1}(1,\bar{2})\mathcal{G}(\bar{2},1')=\delta(1,1'), \tag{B1}$$

where $\delta(1,1')=\delta^3(\mathbf{r}_1-\mathbf{r}_1')\delta(t_1-t_1')$. It can be expressed as

$$\mathcal{G}^{-1}(1,1')=\left[i\frac{\partial}{\partial t_1}+\frac{\hbar}{2m}\nabla_1^2+\frac{\mu}{\hbar}+\frac{2i}{\hbar}v(1,\bar{2})\mathcal{G}(\bar{2},\bar{2}^+)\right]\times\delta(1,1')-\frac{1}{\hbar}\phi(1,1')-\Sigma(1,1'), \tag{B2}$$

with the self energy

$$\Sigma(1,1')=-2^{-1}v(1^+,\bar{2})\chi^{(2)}(\bar{3},1;\bar{2}^+,\bar{2})\mathcal{G}^{-1}(\bar{3},1'). \tag{B3}$$

We take the functional derivative of Eq. (B2) with respect to $\mathcal{G}(2,2')$, multiply the resulting equation by $\chi^{(2)}(2,2';3^+,3)\mathcal{G}(4,1)\mathcal{G}(1',4')$, and integrate it over the indices $1,1',2,2'$; we thus obtain Eq. (16) with the aid of Eq. (15) after some relabeling of indices.

To obtain Eq. (22), we perform functional derivatives of Eq. (B2) successively with respect to $\mathcal{G}(2,2')$ and $\mathcal{G}(3,3')$, multiply the result by $\chi^{(2)}(4^+,4;1,1')\chi^{(2)}(2,2';5^+,5)\chi^{(2)}(3,3';6^+,6)$, and finally integrate it with respect to $1,1',2,2',3,3'$.

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