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Dynamical structure factor of an electron liquid. I

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Electron correlations of an electron liquid at metallic densities are studied by means of a diagrammatic method, the primary aim being the interpretation of the spectral structure observed for several metals in recent inelastic electron and x-ray scattering experiments. Several sum rules and requirements such as the positivity of the pair distribution function, the compressibility sum rule, the frequency moment sum rules, and the conservation law of the local electron number which have hitherto been regarded as guiding principles in theoretical formulation are first examined and discussed in a critical manner. Detailed examinations are made of the proper polarization function within the Hartree-Fock approximation. The lowest-order perturbation expressions giving two-pair excitations and one-pair–one-plasmon excitations are next examined. It is pointed out that these expressions are indeed divergent in the usual one-pair excitation region; the renormalization of the one-particle Green's function is indispensable there. From the above considerations and further discussions, the quasi-one-pair excitation approximation is presented for the purpose of describing the spectral shape in the dynamical structure factor $S(q,\omega)$ in the intermediate wave-number and intermediate frequency regions. In the quasi-one-pair excitation approximation the proper polarization function is constructed from a coherent part of the renormalized one-particle Green's function and a static part of the irreducible particle-hole interaction including strong short-range correlations.

I. INTRODUCTION

Since the 1950's the plasmon excitation in metals has been extensively investigated[1,2] through the measurement of the energy-loss function or the dynamical structure factor $S(q,\omega)$. Recently inelastic electron and x-ray scattering experiments[3–8] on $S(q,\omega)$ for several metals have been extended to the wave-number region beyond the cutoff wave number q_c, with high precision. It has been commonly observed that just before q_c the plasmon dispersion curve bends appreciably from the result in the random-phase approximation (RPA) and its continuation into the one-pair continuum exhibits a very small dispersion up to the wave number much larger than q_c. Moreover, inelastic x-ray scattering experiments[7,8] carried out at Bell Laboratories[7] have revealed that the plasmonlike peak continues up to even the wave number twice as large as the Fermi wave number p_F, showing almost no dispersion or even a negative one in some cases and that on the high-energy side of the plasmonlike peak, a well-developed shoulder or a broad peak arising from individual excitations can be seen. Such a structure in the intermediate wave-number region has been confirmed by Priftis, Boviatsis, and Vradis[8]; the plasmonlike peak and the broad peak have been observed more distinctly for Li. It is generally accepted that those experimental features of $S(q,\omega)$ originate chiefly from dynamical correlation effects of an electron liquid, band-structure effects being of little significance. Those experimental observations have prompted many theoretical studies[9–17] of dynamical correlations in an electron liquid. A theoretical interpretation of the double-peak structure of $S(q,\omega)$ has been the subject of controversy in the last several years. Mukhopadhyay, Kalia, and Singwi[11] have tried to

tackle this problem in a somewhat *ad hoc* way. They have added the imaginary part of the self-energy to the one-electron energy occurring in the Vashishta-Singwi dielectric function. As a consequence they have obtained excitation spectra which have some resemblence to the observed ones. However, their calculated structure in $S(q,\omega)$ could be destroyed when convoluted with an experimental resolution function of width 5 eV. Their work has also been open to question chiefly because it violates the continuity equation. Several authors[12] have attempted to apply Mori's memory-function formalism to this problem without much success. A satisfactory solution has not yet been obtained in spite of such great efforts. But the problem is important, considering the fact that the experiments are expected to reflect some unclarified dynamical effects of electron correlations common to a wide variety of different metals. In this paper and subsequent papers we shall investigate dynamical aspects of electron correlations on the basis of diagrammatic method of perturbation theory. This paper is a detailed report of a previous paper.[18]

The RPA gives an adequate description of long-range correlations such as the plasmon excitation and the screening effect. Its validity, however, is restricted to the high-density region.[2] Short-range correlations which are not included adequately in the RPA are increasingly important as the electron density is lowered to metallic levels. A successful treatment of short-range correlations at metallic densities was first devised by Singwi, Tosi, Land, and Sjölander[19–21] in 1968 (hereafter referred to as STLS theory). Since then, an electron liquid again has actively been studied by means of several methods.[22–29] A better understanding of short-range correlations has been achieved by one of the authors (H.Y.)[26] and others[27–29] by means of the diagrammatic method of perturbation theory. It has been clarified that the particle-particle ladder vertex is indispensable for the description of short-range correlations. On the other hand, the problem of the fine structure in $S(q,\omega)$ with which we are now concerned is much more difficult than that of static correlations such as the pair distribution function $g(r)$. It is very probable that higher-order excitations such as two-pair excitations and one-pair—one-plasmon excitations make a significant contribution to the spectral shape of $S(q,\omega)$ at metallic densities. Careful considerations of such higher-order excitations as well as the particle-particle ladder interactions may be useful for the understanding of dynamical aspects of electron correlations. We shall then pursue the problem consistently from a diagrammatic point of view. We feel that there is important implication in the work by Mukhopadhyay, Kalia, and Singwi in spite of aforementioned shortcomings. In Sec. II we shall first discuss critically several sum rules and requirements which have hitherto been used in theoretical formulation. In Sec. III we shall examine a perturbation calculation of the proper polarization function in the Hartree-Fock approximation and elucidate implications of Hubbard's approximation,[31] which will be useful for further advance calculations. In Sec. IV we shall examine lowest-order perturbation calculations giving two-pair excitations and one-pair—one-plasmon excitations, and point out that they are indeed divergent in the one-pair excitation region. It will be explained that the renormalization of one-particle Green's function is indispensable for the reasonable description of $S(q,\omega)$ in the one-pair excitation region and its neighboring area. With due consideration for such preliminaries in Secs. II—IV we shall obtain, in Sec. V, the quasi-one-pair excitation approximation suitable for describing the spectral shape of $S(q,\omega)$. It will be mentioned that the quasiparticle picture is still useful even for the problem of $S(q,\omega)$ in the intermediate excitation region, if one takes properly into account short-range correlations as well as long-range dynamical correlations in evaluating the energy width of the quasiparticle and the effective particle-hole interaction. Concluding remarks will be given in Sec. VI. The subsequent paper will be devoted to a numerical study. It will be shown there that numerical results are in excellent agreement with experimental observations.

II. SEVERAL SUM RULES AND REQUIREMENTS

Several sum rules and requirements have been considered as guiding principles in developing a theory of electron correlations at metallic densities. We shall criticize whether these guiding principles are suitable to describe dynamical aspects of electron correlations in the intermediate wave-number region. The diagrammatic analyses of their implications are helpful in finding some orientation for a further advanced study. The requirement of $g(r) \geq 0$, the compressibility sum rule, a few frequency moment sum rules, and other requirements such as the conservation law of the local electron number are discussed.

A. Requirement of $g(r) \geq 0$

A clear understanding of the requirement of $g(r) \geq 0$ has been obtained[26–29] by diagrammatic analysis of perturbation processes. The problem of $g(r)$ at short distances is essentially the two-particle scattering problem in the medium. In a paramagnetic electron liquid the pair distribution function $g(r)$ is an arithmetic average of the spin-parallel distribution function $g^{\uparrow\uparrow}(r)$ and the spin-antiparallel one $g^{\uparrow\downarrow}(r)$. Usually $g(r)$ has been calculated with the dielectric formulation where the two functions $g^{\uparrow\uparrow}(r)$ and $g^{\uparrow\downarrow}(r)$ cannot separately be treated. We shall here consider the two functions separately.

Long-range correlations in an electron liquid reduce the Coulomb potential to the screened one. The residual screened potential, however, is still very strong at short distances. In the metallic region where the magnitude of the potential energy is comparable to that of the kinetic energy such a strongly repulsive part of the Coulomb potential makes a significant contribution to electron correlations. In the metallic region a particle-particle ladder type of diagram must be considered up to higher orders to satisfy the requirement $g^{\uparrow\downarrow}(r) \geq 0$. In other words, the wave function of the two electrons with antiparallel spins is deformed at small separations by the short-range part of the Coulomb interaction, and is considerably different from the plane wave.

A reasonable description of $g^{\uparrow\downarrow}(r)$ at short distances is accomplished by calculating polarization functions $P_1^{\uparrow\downarrow}(q,\omega)$ and $P_2^{\uparrow\downarrow}(q,\omega)$ which correspond to time-ordered diagrams shown in Fig. 1:

$$g^{\uparrow\downarrow}(r)=1+\frac{2}{n^2}\int\frac{d\vec{q}}{(2\pi)^3}e^{i\vec{q}\cdot\vec{r}}\left[\int\frac{d\omega}{2\pi i}P_1^{\uparrow\downarrow}(q,\omega)+\int\frac{d\omega}{2\pi i}P_2^{\uparrow\downarrow}(q,\omega)\right] \tag{1}$$

$$=1+\frac{2}{n}\int\frac{d\vec{q}}{(2\pi)^3}e^{i\vec{q}\cdot\vec{r}}[S_1^{\uparrow\downarrow}(q)+S_2^{\uparrow\downarrow}(q)] \tag{2}$$

$$=\frac{4}{n^2}\int\frac{d\vec{p}}{(2\pi)^3}\int\frac{d\vec{p}\,'}{(2\pi)^3}f(p)f(p')\left|1+\int\frac{d\vec{q}}{(2\pi)^3}\frac{[1-f(\vec{p}+\vec{q})][1-f(\vec{p}\,'-\vec{q})]}{\epsilon_p-\epsilon_{\vec{p}+\vec{q}}+\epsilon_{p'}-\epsilon_{\vec{p}\,'-\vec{q}}}I(\vec{p},\vec{p}\,';\vec{q})e^{i\vec{q}\cdot\vec{r}}\right|^2, \tag{3}$$

where n is the electron density, $f(p)$ the Fermi distribution function at zero temperature, $v(q)=4\pi e^2/q^2$, $\epsilon_p=\hbar^2p^2/2m$, and $I(\vec{p},\vec{p}\,';\vec{q})$ is the particle-particle ladder vertex which satisfies the following integral equation:

$$I(\vec{p},\vec{p}\,';\vec{q})=v(q)+\int\frac{d\vec{k}}{(2\pi)^3}v(\vec{q}-\vec{k})\frac{f(p)[1-f(\vec{p}+\vec{k})]f(p')[1-f(\vec{p}\,'-\vec{k})]}{\epsilon_p-\epsilon_{\vec{p}+\vec{k}}+\epsilon_{p'}-\epsilon_{\vec{p}\,'-\vec{k}}}I(\vec{p},\vec{p}\,';\vec{k})\,. \tag{4}$$

The above value of $g^{\uparrow\downarrow}(r)$ is positive for all distances and for any density. The short-distance behavior of $g^{\uparrow\downarrow}(r)$ is significantly affected by the coupling strength of interaction. Its value at zero separation $g^{\uparrow\downarrow}(0)$ is almost 1 for very high densities. On the other hand, $g^{\uparrow\downarrow}(0)$ is about 0.1 for $r_s=4.0$ appropriately to Na.

Let us look at two diagrams $P_1^{\uparrow\downarrow}$ and $P_2^{\uparrow\downarrow}$ in Fig. 1. The diagram for the polarization function $P_1^{\uparrow\downarrow}(q,\omega)$ represents perturbation processes of one-pair excitations. On the other hand, the diagram for $P_2^{\uparrow\downarrow}(q,\omega)$ has two ladder vertices and represents intrinsically processes of two-pair excitations. In order to examine how the above two polarization functions different in character contribute to the short-distance behavior of $g^{\uparrow\downarrow}(r)$, we have estimated each contribution to $g^{\uparrow\downarrow}(0)$ (see Fig. 2). The processes of one-pair excitations make a dominant contribution to $g^{\uparrow\downarrow}(0)$ and keep it positive on the high-density side of the metallic region. At low metallic densities, however, the contribution from the processes of two-pair excitations cannot be neglected to satisfy $g^{\uparrow\downarrow}(r) \geq 0$.

We shall next mention the short-distance behavior of the spin-parallel distribution function $g^{\uparrow\uparrow}(r)$. It vanishes at the origin identically due to the exclusion principle; $g^{\uparrow\uparrow}(0)=0$. This identity can be satisfied by considering each perturbation

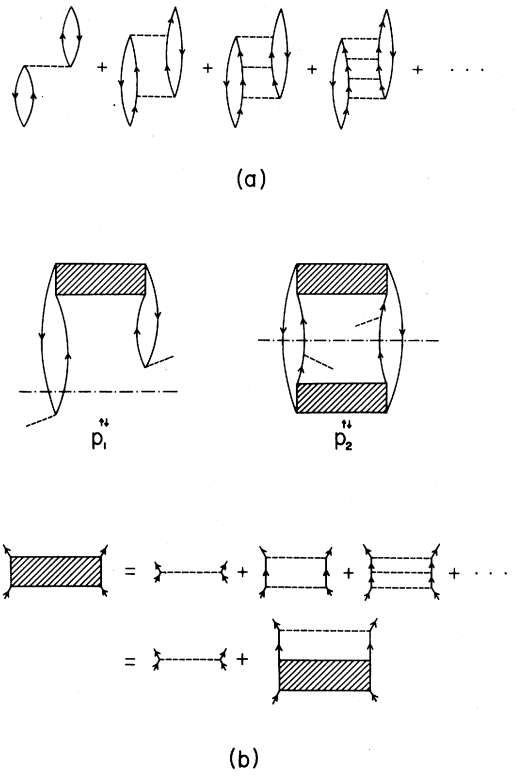


FIG. 1. (a) A series of polarization diagrams which are necessary for the adequate description of the short-distance behavior of $g^{\uparrow\downarrow}(r)$. The solid line with an arrow denotes the free propagator or the noninteracting Green's function. The dotted line denotes the bare Coulomb interaction. (b) Time-ordered representation of the above polarization series, upward lines denote particle states and downward lines hole states. Two types of time-ordered diagrams $P_1^{\uparrow\downarrow}$ and $P_2^{\uparrow\downarrow}$ which contribute to $g^{\uparrow\downarrow}(r)$ and include the particle-particle ladder vertex are shown explicitly. The shaded box denotes the particle-particle ladder vertex which is also shown diagrammatically. Other types of time-ordered diagrams which do not contribute $g^{\uparrow\downarrow}(r)$ or include hole-hole ladder interactions are omitted. The diagram $P_1^{\uparrow\downarrow}$ represents one-pair excitation process and the diagram $P_2^{\uparrow\downarrow}$ two-pair excitation process; the dash-and-dotted horizontal line in each diagram indicates the very energy denominator which represents one-pair excitation or two-pair excitation.

diagram together with its exchange counterpart. The function $g^{\uparrow\uparrow}(r)$ at short distances is little affected by inclusion of the Coulomb interaction owing to the restriction of $g^{\uparrow\uparrow}(0)=0$. If one dares to fulfill $g^{\uparrow\uparrow}(0)=0$ rigorously in the framework of the dielectric formulation, one must choose an exceedingly complicated set of proper polarization diagrams.[30] The dielectric formulation is indispensable for the description of long-range correlations

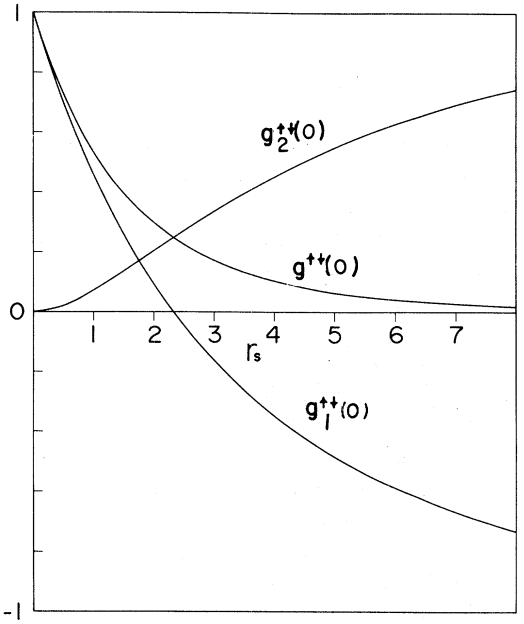


FIG. 2. Values of the spin-antiparallel pair distribution function at the origin $g^{\uparrow\downarrow}(0)$ which are obtained from Eq. (3) are plotted as a function of r_s; r_s is related to the electron density n as $1/n=\frac{4}{3}\pi(r_s a_0)^3$, where a_0 is the Bohr radius. The value of $g^{\uparrow\downarrow}(0)$ is given by the sum of $g_1^{\uparrow\downarrow}(0)$ and $g_2^{\uparrow\downarrow}(0)$ which come from one-pair excitation process and two-pair excitation process, respectively. Values of $g_1^{\uparrow\downarrow}(0)$ and $g_2^{\uparrow\downarrow}(0)$ are also plotted as a function of r_s.

in an electron liquid but it is rather inappropriate for describing short-range correlations.

Finally we remark that Eqs. (2)–(4) lead to the following asymptotic form of $S_1^{\uparrow\downarrow}(q)+S_2^{\uparrow\downarrow}(q)$ for large q:

$$S^{\uparrow\downarrow}(q)=S_1^{\uparrow\downarrow}(q)+S_2^{\uparrow\downarrow}(q) \tag{5}$$

$$=-\frac{4}{3}\frac{\alpha r_s}{\pi}\frac{p_F^4}{q^4}g^{\uparrow\downarrow}(0)+\cdots, \tag{6}$$

$$\alpha=(4/9\pi)^{1/3}.$$

It has also been verified[26,32] that the exact asymptotic form of the static structure factor from spin-antiparallel correlation is given by substituting the exact value of $g^{\uparrow\downarrow}(0)$ into Eq. (6). The contribution from spin-parallel correlation has a higher order asymptotic form for large q. In closing this section, we emphasize that not all diagrammatic processes which are indispensable for $g(r)\geq 0$ for any density can be reduced into the Hubbard type of dielectric function.

B. Compressibility sum rule

The compressibility sum rule implies the following self-consistency. The compressibility estimated directly from the value of the dielectric function in the limit of $\omega=0$, $q\to 0$ is identical with that estimated thermodynamically from the ground-state energy which is calculated with the same dielectric function. Before discussing the sum rule itself at this time we shall indicate an interesting aspect of the STLS theory.[19]

Let us consider an ansatz concerning the two-particle distribution function on which the STLS theory is founded; i.e., the two-particle distribution function in the classical Liouville equation for the system under an external disturbance is replaced by the product of two one-particle distribution functions and the pair distribution function. This replacement is justified rigorously, provided that the external disturbance is stationary in time and has only a spatial Fourier component of the long-wavelength limit; such a special case can substantially be regarded as being in equilibrium. The ansatz requires that the above replacement be also applicable under an arbitrary disturbance. It seems puzzling that the ansatz which might be expected to be appropriate for the description of the long-wavelength component of the density fluctuations leads to a good description of $g(r)$ at short distances. The reason for their success is probably that their self-consistent equation for the local field correction has some resemblance to the integral equation for the particle-particle ladder vertex for large momentum transfers. It is even surprising that the STLS theory happens to give the aforementioned asymptotic form of the static structure factor in which the exact value of $\frac{1}{2}g^{\uparrow\downarrow}(0)$ is replaced by their own self-consistent value of $g(0)$. The STLS theory has been modified[20,21] with the intention of improving[33] the compressibility sum rule but it has given a rather worse behavior of $g(r)$ at short distances.

In the approximation scheme of treating the exchange and correlation potentials statically, Kawazoe, Yasuhara, and Watbe[34] have formulated a self-consistent theory which satisfies rigorously not only the compressibility sum rule but also the spin-susceptibility sum rule. They have obtained numerically self-consistent values of the compressibility and the spin susceptibility as well as $g^{\uparrow\downarrow}(r)$ and $g^{\uparrow\uparrow}(r)$. A fairly good description of $g^{\uparrow\downarrow}(r)$ and $g^{\uparrow\uparrow}(r)$ at short distances has been given, though the two sum rules are not directly related to short-range correlations. It has there been discussed what set of polarization diagrams are needed for the fulfillment of the compressibility sum rule. From the above discussion it may safely be said that their approximated form of the dielectric function involves, in a crude manner, a particle-particle ladder series. Considering the above situation, one may say that the compressibility sum rule is not necessarily a useful guiding principle to the present problem.

C. A few frequency moment sum rules

We shall discuss whether the third and higher frequency moment sum rules are practically useful for describing dynamical aspects of electron correlations in the intermediate wave-number and intermediate frequency regions. The density-density response function $\chi(q,\omega)$ is related to the dielectric function as

$$\chi(q,\omega)=\frac{1}{v(q)}\left[\frac{-1}{\epsilon(q,\omega)}+1\right]. \tag{7}$$

For the investigation of the high-frequency behavior of $\chi(q,\omega)$, the following formal expression has conventionally been used:

$$\chi(q,\omega)=\frac{1}{i\hbar}\int_{-\infty}^{\infty}d\omega\langle[\rho_q(t),\rho_q^{\dagger}(0)]\rangle e^{i\omega t}\Theta(t) \tag{8}$$

$$=\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\frac{\mathrm{Im}\chi(q,\omega')}{\omega'-\omega-i\delta}\quad(\delta\to 0+) \tag{9}$$

$$=-\frac{M_1(q)}{\omega^2}-\frac{M_3(q)}{\omega^4}-O(\omega^{-6}), \tag{10}$$

where $\Theta(x)=1$ for $x>0$, zero otherwise, $\rho_q(t)$ is the density fluctuation operator in the Heisenberg representation, and $\langle\ \rangle$ denotes the expectation value with respect to the ground state. The expansion coefficients are formally written as

$$M_{2l-1}(q)=\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\omega^{2l-1}\mathrm{Im}\chi(q,\omega). \tag{11}$$

The first moment $M_1(q)$ means the well-known f-sum rule

$$M_1(q)=\frac{nq^2}{m}, \tag{12}$$

where m is the electron mass and n the electron density. The f-sum rule must, of course, be satisfied, if one studies the spectral shape of $S(q,\omega)$ at all. The third frequency moment $M_3(q)$ can be seen in the literature[35–37] as well. Several authors

have attempted[12,14,16,36,37] to formulate their theories in such a way that the third frequency moment sum rule is satisfied. This type of theory may give fairly well the plasmon dispersion for very long wavelengths. But it cannot explain the anomalous behavior of the plasmon dispersion around q_c.

The above high-frequency expansion of $\chi(q,\omega)$ has been considered as if it might be possible up to the infinite order for an arbitrary system. The following fact, however, is worthy of particular attention. An asymptotic form of $\mathrm{Im}\chi(q,\omega)$ for a degenerate electron liquid is given by Glick and Long[39] as follows;

$$\mathrm{Im}\chi(q,\omega)=v(q)^{-1}\frac{92}{135}\frac{p_F^6}{\pi^2 a_0^3}\frac{q^2}{(m\omega)^{11/2}},\quad q\lesssim p_F,\quad \omega>>\epsilon_F \tag{13}$$

where ϵ_F is the Fermi energy. It arises from two-pair excitations of second-order perturbation. Let us substitute the above asymptotic form into the fifth moment integral. The integral is quite divergent; the spectral function $\mathrm{Im}\chi(q,\omega)$ is positive definite for $\omega>0$ and an even function with respect to ω,

$$\int_{\omega_c}^{\infty} d\omega\,\omega^5\omega^{-11/2}\to\infty\ , \tag{14}$$

where ω_c is the minimum value of ω for which Eq. (13) holds with sufficient accuracy. The divergence of the fifth moment implies that the formal expansion in the squared inverse powers of ω is not valid for higher orders; a term of order lower than ω^{-6} should follow immediately that of order ω^{-4}. To obtain the correct term, we shall substitute the above asymptotic form into the spectral representation of $\chi(q,\omega)$ given by Eq. (9) and first perform the integration over a high-frequency region where the above asymptotic form holds. A term of order $\omega^{-11/2}$ is then obtained from a revised expansion of $\chi(q,\omega)$; that is, in the correct expansion the real part of $\chi(q,\omega)$ has also a term of order $\omega^{-11/2}$. In other words, the $t\gtrsim 0$, the function $\chi(q,t)$ which is the Fourier transform of $\chi(q,\omega)$ can be differentiated with respect to t up to three times but it cannot be done five times. In conclusion, the higher moment expansion of $\chi(q,\omega)$ are not expected to be helpful in studying electron correlations in the intermediate wave-number and intermediate frequency regions.

D. A few conservation laws

A self-consistent method which guarantees conservation of the local electron number, local momentum, and local energy has been presented by Baym and Kadanoff.[40] It has been elucidated what set of polarization diagrams should be chosen for that purpose. The fulfillment of such local conservation laws is essential for the description of transport phenomena. The shielded interaction approximation[40] has been proposed as one of such self-consistent approximations for an electron liquid. We remark here that the shielded interaction approximation does not satisfy the requirement of $g(r)\geq 0$ at metallic densities. It is desirable that a theory valid at metallic densities could be developed according to the self-consistent scheme. However, such calculations are formidable because they are exceedingly complicated.

The conservation laws of the local electron number as well as the third frequency moment sum rule can also be maintained easily, if one resorts to Mori's memory-function formalism.[38] This formalism has been employed[12] for an electron liquid, without much success. We may consequently say that the fulfillment of the conservation of the local electron number[14,41] is of secondary importance, when one is interested in electron correlations in the intermediate wave-number and intermediate frequency regions where typical values of wave number and frequency are characterized by the Fermi wave number and the Fermi energy or the plasma frequency, respectively.

III. HARTREE-FOCK APPROXIMATION

In this and subsequent sections we shall give a few illustrations of perturbation calculations for the polarization function. A few difficulties involved therein will be elucidated. Adequate consideration of these difficulties is very helpful for obtaining the quasi-one-pair excitation approximation which will be presented in Sec. V. We shall begin with formal equipment. The dielectric function $\epsilon(q,\omega)$ is written[42,43] in terms of the proper

polarization function $\pi(q,\omega)$ as

$$\epsilon(q,\omega)=1+v(q)\pi(q,\omega)\ . \tag{15}$$

The dynamical structure factor $S(q,\omega)$ for $\omega\geq 0$ is related to the imaginary part of the inverse dielectric function as

$$S(q,\omega)=-[\pi v(q)]\mathrm{Im}^{-1}[1/\epsilon(q,\omega)]\ . \tag{16}$$

The proper polarization function is formally written[42,43] in terms of the one-particle Green's function $G(p,\epsilon)$ and the proper vertex function $\tilde{\Lambda}(\vec{p},\epsilon;\vec{q},\omega)$,

$$\pi(q,\omega)=-2\int\frac{d\epsilon}{2\pi i}\int\frac{d\vec{p}}{(2\pi)^3}G(p,\epsilon)G(\vec{p}+\vec{q},\epsilon+\omega)\tilde{\Lambda}(\vec{p},\epsilon;\vec{q},\omega)\ . \tag{17}$$

Here, the proper vertex function $\tilde{\Lambda}(\vec{p},\epsilon;\vec{q},\omega)$ is the solution of the following integral equation:

$$\tilde{\Lambda}(\vec{p},\epsilon;\vec{q},\omega)=1+\int\frac{d\epsilon'}{2\pi i}\int\frac{d\vec{p}\,'}{(2\pi)^3}\tilde{I}(\vec{p},\epsilon;\vec{p}\,',\epsilon';\vec{q}\omega)G(p',\epsilon')G(\vec{p}\,'+\vec{q},\epsilon'+\omega)\tilde{\Lambda}(\vec{p}\,',\epsilon';\vec{q},\omega)\ , \tag{18}$$

where $\tilde{I}$ is the irreducible particle-hole interaction and $G(p,\epsilon)$ includes its self-energy part $\Sigma(p,\epsilon)$ as

$$G(p,\epsilon)=\frac{1}{\epsilon-\epsilon_p-\Sigma(p,\epsilon)}\ . \tag{19}$$

We shall first examine the Hartree-Fock approximation, which will be a useful preliminary to further advanced approximations. If one approximates $\tilde{I}$ and Σ in Eqs. (18) and (19) to lowest order in the Coulomb interaction, i.e.,

$$\tilde{I}(\vec{p}_1,\epsilon_1;\vec{p}_2,\epsilon_2;\vec{q},\omega)\rightarrow v(p_1-\vec{p}_2),\quad \Sigma(p,\epsilon)\rightarrow\Sigma^{\mathrm{HF}}(p)=-\int\frac{d\vec{p}\,'}{(2\pi)^3}v(\vec{p}-\vec{p}\,')f(\vec{p}\,')\ ,$$

then the Hartree-Fock expression for $\pi(q,\omega)$ results,

$$\pi^{\mathrm{HF}}(q,\omega)=-2\int\frac{d\vec{p}_1}{(2\pi)^3}\left[\frac{f(p_1)[1-f(\vec{p}_1+\vec{q})]}{\omega+i\delta+E(p_1)-E(\vec{p}_1+\vec{q})}-\frac{f(\vec{p}_1+\vec{q})[1-f(p_1)]}{\omega-i\delta+E(p_1)-E(\vec{p}_1+\vec{q})}\right]\tilde{\Lambda}^{\mathrm{HF}}(\vec{p}_1;\vec{q},\omega)\ , \tag{20}$$

$$\tilde{\Lambda}^{\mathrm{HF}}(\vec{p}_1;\vec{q},\omega)=1+\int\frac{d\vec{p}_2}{(2\pi)^3}v(\vec{p}_1-\vec{p}_2)\left[\frac{f(p_2)[1-f(\vec{p}_2+\vec{q})]}{\omega+i\delta+E(p_2)-E(\vec{p}_2+\vec{q})}-\frac{f(\vec{p}_2+\vec{q})[1-f(p_2)]}{\omega-i\delta+E(p_2)-E(\vec{p}_2+\vec{q})}\right]\times\tilde{\Lambda}^{\mathrm{HF}}(\vec{p}_2;\vec{q},\omega)\ , \tag{21}$$

where the energy spectrum $E(p)$ is shifted from ϵ_p by an amount of the Hartree-Fock self-energy. In this approximation a particle-hole ladder series of interactions shown in Fig. 3 are summed. It is straightforward to show that the imaginary part of $\pi^{\mathrm{HF}}(q,\omega)$ for $\omega>0$ is written as

$$\mathrm{Im}\pi^{\mathrm{HF}}(q,\omega)=2\pi\int\frac{d\vec{p}}{(2\pi)^3}\left|\tilde{\Lambda}^{\mathrm{HF}}(\vec{p}+\vec{q};\vec{q},\omega)\right|^2 f(p)[1-f(\vec{p}+\vec{q})]\delta(\omega+E(p)-E(\vec{p}+\vec{q}))\ . \tag{22}$$

Obviously it involves only one-pair excitations, though its frequency boundaries of one-pair excitations are shifted from the RPA ones by the self-energy contributions.

We shall discuss here in some detail Hubbard's approximation to the Hartree-Fock expression, since its underlying idea may be helpful when we treat higher-order corrections. Hubbard has arrived[31] at his approximation in an attempt to allow for exchange corrections to a RPA series of improper polarization diagrams. If one replaces the interaction $v(\vec{p}_1-\vec{p}_2)$ on the right-hand side of Eq. (21) by its averaged value within Fermi spheres proposed by Hubbard,

$$\langle v(\vec{p}_1-\vec{p}_2)\rangle_{\vec{p}_1\vec{p}_2+\vec{q}}\simeq 4\pi e^2/(q^2+p_F^2)$$

and the energy spectrum $E(p)$ by ϵ_p, the resulting expression for $\pi(q,\omega)$ amounts to Hubbard's approximation. To make clear physical implications of his approximation we shall substitute into Eq. (20) the first iterative solution for $\tilde{\Lambda}^{\mathrm{HF}}$ which is obtained by putting $\tilde{\Lambda}^{\mathrm{HF}}=1$ in the second term on the right-hand side of Eq. (21). Then the four

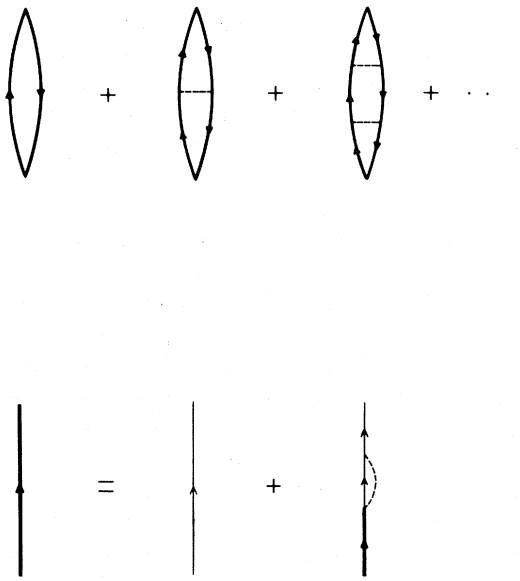


FIG. 3. Hartree-Fock series of polarization diagrams. The bold line denotes the Hartree-Fock Green's function.

separate terms written in a product of two energy denominators appear. Each of these four terms has a different set of Fermi distribution functions. So far as one is concerned with those two terms which have poles on both sides of the real axis, an approximate estimation of the interaction $v(\vec{p}_1-\vec{p}_2)$ proposed by Hubbard may be reasonable; it can be seen from

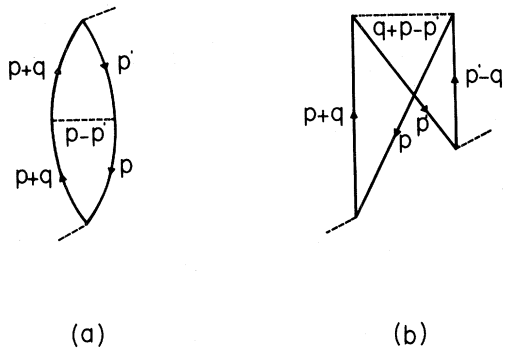
$$f(p_1)f(\vec{p}_1+\vec{q})[1-f(\vec{p}_1+\vec{q})][1-f(p_2)]$$

or

$$f(\vec{p}_1+\vec{q})f(p_2)[1-f(p_1)][1-f(\vec{p}_2+\vec{q})]$$

in the two terms that the magnitude of $\vec{p}_1-\vec{p}_2$ is proportional to that of q for sufficiently large q. The above two terms make a contribution to the static structure factor $S(q)$ or the total energy (see Fig. 4). On the other hand, the other two terms which have two poles on either side of the real axis vanish when integrated over frequencies and then make no contribution to the total energy.[44,45] Hubbard's expression is thus useful for estimating the second-order exchange energy and higher-order contributions in an approximate manner.

A more elaborate approximation for $\pi^{HF}(q,\omega)$ has been proposed by Kleinmann[44] and Langreth[45] in their study of the dielectric function. For the interaction $v(\vec{p}_1-\vec{p}_2)$ entering in those two terms which do not contribute to the total energy they



(a) (b)

FIG. 4. Lowest-order Hartree-Fock diagrams in the time-ordered representation. Diagram (a) does not contribute to the static structure factor $S(q)$. Diagram (b) contributes to $S(q)$.

have properly chosen a different approximate form, considering that the magnitude of $\vec{p}_1-\vec{p}_2$ in such two terms is independent of q. But it has been found thereby that in order to satisfy the f-sum rule the elaborate approximation to the vertex correction requires explicit inclusion on the self-energy shift. From the discussion of the f-sum rule one may consequently say that Hubbard's expression is devised in such a manner that cancellation between the self-energy shift and a fraction of the vertex correction is, from the first, taken into account; in Hubbard's treatment of the vertex correction, the explicit inclusion of the self-energy shift is not needed. Such a treatment of the cancellation will also be applicable practically to an approximate calculation of higher-order polarization functions.

A numerical estimation of the imaginary part of the exact Hartree-Fock expression has been performed by Woo and Jha.[46] It exhibits a spectral shape similar to Hubbard's. One may then say that the treatment of the vertex correction and the self-energy shift after Hubbard's manner is fairly good for the description of the spectral shape of $\mathrm{Im}\pi(q,\omega)$. No interesting structure has, however, been found in the resulting $\mathrm{Im}\pi(q,\omega)$ within the Hartree-Fock approximation except that its spectrum as a whole is, by some amount, shifted to the low-energy side, compared with the RPA case. Such a shift is due to the attractive Coulomb interaction between particles and holes.

Very recently several authors have calculated[15,47] the lowest-order perturbation terms for the proper polarization function. They have then found that the spectrum of the imaginary part of such polarization corrections exhibits singular behaviors at frequency boundaries of one-pair excitations. This

type of singularities could, in principle, be removed by renormalizing the one-particle Green's function within the framework of the Hartree-Fock approximation.

IV. TWO PAIR AND ONE-PAIR—ONE-PLASMON EXCITATIONS

If one proceeds to a further advanced stage of approximation,[48] the situation is further complicated by the fact that vertex and self-energy corrections beyond the Hartree-Fock approximation depend on the frequency. The existence of higher-order excitations such as two-pair excitations and one-pair—-one-plasmon excitations originates from imginary parts of vertex and self-energy corrections.

We shall examine in some detail the lowest-order perturbation calculations which give two-pair excitations and one-pair—one-plasmon excitations. It should be noted that in the usual one-pair excitation region these perturbation calculations result in difficulties of divergence, owing to a piling up of the same energy denominator. Only when q and ω are situated far away from the one-pair excitation region do these perturbation expressions have significance. Such calculations have practically been performed[48–51] to estimate a damping of the plasmon in the low-wavelength region. The lowest-order diagrams which contribute to two-pair excitations are shown in Figs. 5(a)—5(c). There are also exchange counterparts which are not shown in Fig. 5. It is, however, sufficient for our aim to take the direct diagrams alone. Let us first consider the polarization functions corresponding to Figs. 5(a) and 5(b). The formal expression for those imaginary parts which come from two-pair exciting parts in these diagrams can be written as

$$\mathrm{Im}\pi^{(a)+(b)}(q,\omega)=4\pi\int\frac{d\vec{k}_1}{(2\pi)^3}\int\frac{d\vec{k}_2}{(2\pi)^3}f(\vec{k}_1-\vec{k}_2)[1-f(k_1)]f(\vec{k}_3-\vec{q})[1-f(\vec{k}_3-\vec{k}_2)]$$
$$\times v(k_2)^2\left[\frac{1}{\omega+\epsilon_{\vec{k}_3-\vec{q}}-\epsilon_{k_3}}-\frac{1}{\omega+\epsilon_{\vec{k}_3-\vec{k}_2-\vec{q}}-\epsilon_{\vec{k}_3-\vec{k}_2}}\right]^2$$
$$\times\delta(\omega+\epsilon_{\vec{k}_1-\vec{k}_2}-\epsilon_{k_1}+\epsilon_{\vec{k}_3-\vec{q}}-\epsilon_{\vec{k}_3-\vec{k}_2})\,. \tag{23}$$

It is straightforward to see from Eq. (23) that two-pair excitations are formally possible for all frequencies if $q\leq 4p_F$. Let us expand the square of the term in large parentheses in Eq. (23). The resulting two squared denominators come from the contribution of the self-energy type corresponding to diagram (a) in Fig. 5. The cross terms of two denominators in the large parentheses, on the other hand, come from that of the vertex type corresonding to diagram (b).

Next, we shall include the contribution corresponding to diagram (c) of the vertex type which are composed of two bubbles. Then, the total contribution can be written as

$$\mathrm{Im}\pi^{(a)+(b)+(c)}(q,\omega)=2\pi\int\frac{d\vec{k}_1}{(2\pi)^3}\int\frac{d\vec{k}_2}{(2\pi)^3}\int\frac{d\vec{k}_3}{(2\pi)^3}f(\vec{k}_1-\vec{k}_2)[1-f(k_1)]f(\vec{k}_3-\vec{q})[1-f(\vec{k}_3-\vec{k}_2)]$$
$$\times\left[v(k_2)\left[\frac{1}{\omega+\epsilon_{\vec{k}_3-\vec{k}_2-\vec{q}}-\epsilon_{\vec{k}_3-\vec{k}_2}}-\frac{1}{\omega+\epsilon_{\vec{k}_3-\vec{q}}-\epsilon_{k_3}}\right]\right.$$
$$\left.+v(k_2-q)\left[\frac{1}{\omega+\epsilon_{\vec{k}_1-\vec{q}}-\epsilon_{k_1}}-\frac{1}{\omega+\epsilon_{\vec{k}_1-\vec{k}_2}-\epsilon_{\vec{k}_1-\vec{k}_2+\vec{q}}}\right]\right]^2$$
$$\times\delta(\omega+\epsilon_{\vec{k}_1-\vec{k}_2}-\epsilon_{k_1}+\epsilon_{\vec{k}_3-\vec{q}}-\epsilon_{\vec{k}_3-\vec{k}_2})\,. \tag{24}$$

These cross terms of the two terms in large parentheses, which are obtained by expanding the square of the term in square brackets in Eq. (24), are the contribution corresponding to diagram (c). In passing we shall remark here upon the following fact. Each contribution to the static structure factor $S(q)$ from the two diagrams (a) and (b) or that from diagram (c) is of order q^2 in the limit of small q. However, they cancel each other completely. The net contribution then becomes of order q^4, as is expected for multipair contributions.[2]

A similar situation is found when one calculates the lowest-order contribution of one-pair—one-plasmon excitations. The lowest-order diagrams which contribute to one-pair—one-plasmon excitatons are shown in Fig. 6; they are also classified into the self-energy type (a) and the vertex one (b). The formal expression for imaginary parts which come from one-pair—one-plasmon exciting parts in these diagrams can be written as

$$\mathrm{Im}\pi(q,\omega)=2\pi\int\frac{d\vec{p}}{(2\pi)^3}\int\frac{d\vec{k}}{(2\pi)^3}\frac{\omega_{\mathrm{pl}}(0)^2}{2\omega_{\mathrm{pl}}(k)}v(k)\Theta(k_c-k)f(p)[1-f(\vec{p}+\vec{q}+\vec{k})]$$
$$\times\left[\frac{1}{\omega+\epsilon_p-\epsilon_{\vec{p}+\vec{q}}}-\frac{1}{\omega+\epsilon_{\vec{p}+\vec{q}}-\epsilon_{\vec{p}+\vec{q}+\vec{k}}}\right]^2$$
$$\times\delta(\omega+\epsilon_p-\epsilon_{\vec{p}+\vec{q}+\vec{k}}-\omega_{\mathrm{pl}}(k))\,, \tag{25}$$

where $\omega_{\mathrm{pl}}(k)$ denotes the plasmon dispersion and k_c is the cutoff wave number. It is straightforward to show from Eq. (25) that the threshold frequency of one-pair—one-plasmon excitations is the plasma frequency $\omega_{\mathrm{pl}}(0)$ for $q\leq 2p_F$ and is $\omega_{\mathrm{pl}}(q-2p_F)$ for q slightly larger than $2p_F$. In a similar way as the case for two-pair excitations, those two squared denominators, which are obtained by expanding the square of the term in large parentheses in Eq. (25), come from the self-energy contribution, and the cross terms of two denominators in the large parentheses from the vertex contribution.

The following fact is worthy of more recognition. The above perturbation expressions become physically inadequate as the frequency variable ω approaches the one-pair excitation region with the wave number q fixed. They increase extraordinarily in the immediate vicinity of the one-pair excitation region and are quite divergent when ω is within the one-pair excitation region. The reason is that the argument of the δ function in Eqs. (24) or (25) and both or one of the squared denominators in it, originating from the correction of the self-energy type, can be equal to each other for frequencies in the one-pair excitation region.

Glick and Long[39] have practically estimated the lowest-order perturbation terms of two-pair excitations for $q\lesssim p_F$, resorting to the Monte Carlo method. Their numerical estimation of $\mathrm{Im}\epsilon(q,\omega)$ gives a reasonable value around the plasmon frequency, but it exhibits an extraordinarily large value when ω approaches the immediate vicinity of the one-pair excitation region, due to the above-mentioned fact. Hermanson[52] has also calculated

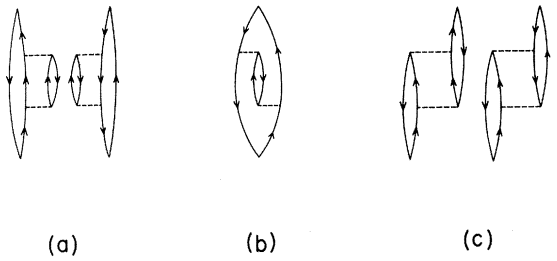


FIG. 5. Lowest-order diagrams contributing to two-pair excitations. (a) Self-energy type of diagrams. (b) Vertex type of diagram. (c) The other vertex type of diagrams consisting of two bubbles.

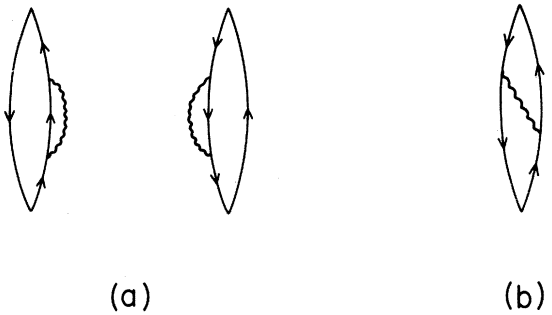


FIG. 6. Lowest-order diagrams contributing to one-pair—one-plasmon excitations. The wavy line denotes the plasmon propagator. (a) Self-energy type of diagrams. (b) Vertex type of diagram.

the lowest-order perturbation terms of one-pair—one-plasmon excitations in his study of the optical-absorption strength of alkali metals in the ultraviolet region. Though he did not try to estimate numerically his expression, it is indeed divergent in the frequency region of his interest.

For the reasonable evaluation of $S(q,\omega)$ in the one-pair excitation region and its neighboring area, one must therefore renormalize the one-particle Green's function; thereby self-energy corrections can be taken into account up to infinite order. Those perturbation terms which involve a piling up of the same energy denominator can thus be summed into a convergent form. Once the one-particle Green's function is renormalized, it is no longer possible to separate, in a distinct way, two-pair excitations or one-pair—one-plasmon excitations from the one-pair ones. It may be expected that in real metals there occurs actually a strong coupling between the usual one-pair excitations and two-pair excitations or one-pair—one-plasmon excitations.

V. QUASI-ONE-PAIR EXCITATION APPROXIMATION

In the light of the knowledge obtained in Secs. II—IV we shall here present the quasi-one-pair excitation approximation which will be suitable for describing the spectral structure in $S(q,\omega)$. Hubbard's expression for the dielectric function is originally a simplified version of the Hartree-Fock one. This type of dielectric function involving only one-pair excitations has been used extensively with the intention of allowing for higher-order corrections as well. Judging from the diagrammatic point of view, dielectric functions calculated in advanced approximations necessarily involve not only one-pair excitations but also two-pair excitations, one-pair—one-plasmon excitations, etc. The above simplified manner of treating correlation corrections may somehow be tolerable, so far as such a type of dielectric function is employed for the purpose of obtaining the knowledge of the static structure factor $S(q)$ or the pair distribution function $g(r)$, where one is concerned with not fine details of the spectral structure in $S(q,\omega)$ but its integrated value over all frequencies. It is, however, very likely that some type of perturbation processes which do not appreciably affect the behavior of $g(r)$ or $S(q)$ may be responsible for the fine details of the spectral structure.

A faithful perturbation calculation of the dielectric function is an exceedingly difficult task, involving a very delicate interference between vertex and self-energy corrections and is beyond the scope of the practical treatment. Instead, we shall proceed based on the knowledge obtained in previous sections and further physical considerations.

The plasmon excitation is well-defined and exhausts almost all oscillator strengths in the long-wavelength region. Then, even in the intermediate wave-number region those perturbation processes which represent virtual plasmon emissions and absorptions are expected to play a significant role. The particle-particle ladder vertex is also indispensable in such a wave-number region. We may then anticipate that the characteristic structure in $S(q,\omega)$ is closely related to the above two effects. Even such perturbation processes are still too complicated to be treated rigorously in terms of the complete form of the renormalized one-particle Green's function and the frequency-dependent particle-hole interaction. Instead, we shall consider how a sharp frequency dependence of $S(q,\omega)$ is made up and manage to allow for these physical processes.

We shall first rewrite the proper polarization function given by Eqs. (17) and (18) using the iterative solution for $\tilde{\Lambda}$:

$$\pi(q,\omega)=-2\int\frac{d\epsilon}{2\pi i}\int\frac{d\vec{p}}{(2\pi)^3}G(p,\epsilon)G(\vec{p}+\vec{q},\epsilon+\omega)\tilde{\Lambda}(\vec{p},\epsilon;\vec{q}\omega) \tag{26}$$

$$=-2\int\frac{d\epsilon}{2\pi i}\int\frac{d\vec{p}}{(2\pi)^3}\Big[G(p,\epsilon)G(\vec{p}+\vec{q},\epsilon+\omega)$$

$$+G(p,\epsilon)G(\vec{p}+\vec{q},\epsilon+\omega)\int\frac{d\epsilon'}{2\pi i}\int\frac{d\vec{p}\,'}{(2\pi)^3}\tilde{I}(\vec{p},\epsilon;\vec{p}\,',\epsilon';\vec{q},\omega)$$

$$\times G(p',\epsilon')G(\vec{p}\,'+\vec{q},\epsilon'+\omega)+\cdots\Big]. \tag{27}$$

Let us split[42] the renormalized one-particle Green's function into two parts; one is a coherent part which ar-

ises from the contribution of a quasiparticle's (or a quasihole's) pole $[\epsilon=E(p)=\epsilon_p+\Sigma(p,E(p))]$ on the analytically continued plane. The other is an incoherent part arising from configurations of several elementary excitations:

$$G(p,\epsilon)=\frac{z_p}{\epsilon-\epsilon_p-\Sigma_1(p,E(p))-i\Sigma_2(p,E(p))}+G^{\rm inc}(p,\epsilon)\ , \tag{28}$$

where z_p is the renormalization constant of the quasiparticle. The real part of the self-energy correction $\Sigma_1(p,E(p))$ in Eq. (28) is the energy shift of the quasiparticle. The imaginary part $\Sigma_2(p,E(p))$ gives its energy width. A form[53,54] of the energy width $(p-p_F)^2$ which is valid in the immediate neighborhood of p_F is owing to the Pauli principle and the screening effect. The notation of a quasiparticle might often be supposed to work well only in such a confined region. It should, however, be noted that the notion of a quasiparticle is founded on short-range correlations as well as the long-range screening effect. Intuitively speaking, the quasiparticle consists of an electron and its surrounding correlation hole. In the metallic density region the magnitude of $\Sigma_2(p,E(p))$ for p larger than p_F may be appreciably smaller than that expected from the RPA, since short-range correlations weaken substantially large momentum transfer part of the Coulomb interaction. Even if the magnitude of the energy width itself is not so small, the quasiparticle picture may still be tolerable so far as the energy measured from the Fermi level is large enough, compared with its energy width. We shall then employ the coherent part of $G(p,\epsilon)$ for the problem related to intermediate excitations.

Now let us return to Eq. (26). For the moment we shall set $\tilde{\Lambda}=1$ there. A sharp frequency dependence of that term does arise from a convolution integral of two coherent parts of renormalized Green's functions. On the other hand, the remaining convolution integrals including the incoherent part $G^{\rm inc}(p,\epsilon)$ are not expected to give any significant frequency dependence. At first sight one might suppose that the magnitude of the coherent contribution is considerably reduced owing to the appearance of renormalizaton constants. For example, the value of z_p estimated[54] to first order in the RPA dynamically screened interaction is about 0.77 at $p=p_F$ for $r_s=2.0$; note that this reduction effect originates, for the most part, from the process emitting plasmon virtually. It has, however, been argued by several authors[52,55,56] that the above GG approximation is seriously unbalanced and that the renormalization effect should be considered together with vertex corrections, since there are very large cancellations between the two effects. In a related problem[55,56] of many-body effects on the optical-absorption strength in the interband region for alkali metals, it has been indicated that the above renormalization effect is overcome by inclusion of the proper vertex function $\tilde{\Lambda}$. The enhancement of one-pair absorption strength due to many-body effects was estimated at its threshold frequency to first-order approximation in the RPA dynamically screened interaction. The vertex correction is considerably large but it is, for the most part, canceled by the self-energy correction including the renormalization effect. The net effect on the strength of one-pair absorption is rather small and amounts to the vertex correction calculated in the *statically* screened interaction. In the present problem we may also anticipate that those dynamical parts of the irreducible particle-hole interaction $\tilde{I}$ which are caused by virtual emission and absorption of plasmons cancel the renormalization effect and that the net effect of $\tilde{I}$ can be evaluated using the statically screened interaction. As a consequence, we neglect, from the first, the renormalization effect together with the above dynamical effect of the irreducible particle-hole interaction.

Let us consider the second term on the right-hand side of Eq. (27). Suppose that every coherent part of four Green's functions in it corresponds to the particle state with its pole on the lower half-plane. If the particle-hole interaction is assumed to be independent of frequency variables, the term vanishes. It should be remembered that the very frequency dependence of $\tilde{I}$ can cause the term to survive even in such a case. A significant part of its frequency dependence which comes from the virtual-plasmon excitation is canceled by the renormalizing effect from the self-energy. The remaining part of frequency dependence has a rather inconspicuous effect. An example of perturbation processes which can survive owing to the remaining part of frequency dependence is diagrammatically represented in Fig. 1(b) in Sec. II; see diagram $P_2^{\uparrow\downarrow}$ which is needed for guaranteeing $g^{\uparrow\downarrow}(r)\geq 0$ in the low metallic region. In general, those contribu-

tions which can survive only with the help of the frequency dependence of $\tilde{I}$, such as the above-mentioned example, constitute a broad background because of destructive interference.

After all, it is found from Eqs. (17) and (18) that the proper polarization function in our quasi-one-pair excitation approximation can be written as

$$\pi(q,\omega)=\frac{\tilde{\pi}^{(0)}(q,\omega)}{1+\tilde{I}(q)\tilde{\pi}^{(0)}(q,\omega)}\ , \tag{29}$$

where $\tilde{\pi}^{(0)}(q,\omega)$ is the free polarization function with the modification of the energy width of the quasiparticle $\Gamma(p)$:

$$\tilde{\pi}^{(0)}(q,\omega)=-2\int\frac{d\vec{p}}{(2\pi)^3}\left[\frac{f(p)[1-f(\vec{p}+\vec{q})]}{\omega+i[\Gamma(p)-\Gamma(\vec{p}+\vec{q})]+\epsilon_p-\epsilon_{\vec{p}+\vec{q}}}-\frac{f(\vec{p}+\vec{q})[1-f(p)]}{\omega+i[\Gamma(\vec{p}+\vec{q})-\Gamma(p)]+\epsilon_p-\epsilon_{\vec{p}+\vec{q}}}\right]. \tag{30}$$

The problem is thus to evaluate the energy width of the quasiparticle and the appropriately averaged form of the static particle-hole interaction $\tilde{I}(q)$. A sharp frequency dependence of $\mathrm{Im}\pi(q,\omega)$ in the one-pair excitation region and its neighboring area is expected to be represented by a coherent contribution from each pair of GG separated by a static particle-hole interaction $\tilde{I}(q)$ in the expansion of Eq. (27).

For the quantitative estimation of the particle-hole interaction at metallic densities we must allow for short-range correlations. That is, those parts of the statically screened interaction whose momentum transfers are larger than the order of $\hbar p_F$ are considerably reduced by inclusion of the particle-particle ladder vertex. In the same spirit as Hubbard's explained in Sec. III we shall here treat particle-hole interactions and the self-energy shift. That is, the cancellation between the self-energy shift and a fraction of the vertex correction is, from the first, taken into account; a form of $\tilde{I}(q)$ appropriate for those types of polarization processes which contribute to the static structure factor or the total energy is commonly used. The omission of the self-energy shift can also be supported by the fact that it is almost independent of the magnitude of the wave number.[54] The irreducible particle-hole interaction in our approximation is given by the use of the particle-particle ladder vertex. The irreducible particle-hole interaction is originally a quantity dependent of two internal wave numbers $\vec{p}$, $\vec{p}\,'$ smaller than p_F. It may safely be approximated by its averaged value over $\vec{p}$ and $\vec{p}\,'$ within Fermi spheres:

$$\tilde{I}(q)=-G(q)v(q)=\tfrac{1}{2}\langle I(\vec{p},\vec{p}\,';\vec{q})-v(q)\rangle_{\vec{p}\,\vec{p}\,'}+\tfrac{1}{2}\langle I(\vec{p},\vec{p}\,';\vec{q})-v(q)-I(\vec{p},\vec{p}\,';\vec{p}-\vec{p}\,'+\vec{q})\rangle_{\vec{p}\,\vec{p}\,'}\ , \tag{31}$$

where the angular bracket $\langle\ \rangle_{\vec{p}\,\vec{p}\,'}$ denotes an averaged value over $\vec{p}$ and $\vec{p}\,'$ within Fermi spheres. The function $G(q)$ is the so-called local-field correction. The first term on the right-hand side of Eq. (31) arises from spin-antiparallel correlation. The second term, on the other hand, arises from spin-parallel correlation; it consists of the direct term

$$\tfrac{1}{2}[\langle I(\vec{p},\vec{p}\,';\vec{q})\rangle_{\vec{p}\,\vec{p}\,'}-v(q)]$$

and the exchange term

$$-\tfrac{1}{2}\langle I(\vec{p},\vec{p}\,';\vec{q}+\vec{p}-\vec{p}\,')\rangle_{\vec{p}\,\vec{p}\,'}\ .$$

The particle-particle ladder vertex is originally intended for the description in the region $q\gtrsim p_F$ but it must be screened for small q, when necessary. In lowest order Eq. (31) is reduced to

$$-\tfrac{1}{2}\langle v(\vec{q}+\vec{p}-\vec{p}\,')\rangle_{\vec{p}\,\vec{p}\,'}\ ,$$

which is the exchange correction discussed in Sec. III.

The energy width of the quasiparticle can be represented by the imaginary part of the self-energy evaluated at the unperturbed pole. For its quantitative estimation[54] at metallic densities, it is necessary to allow for short-range correlations arising from the exchange effect and the Coulomb repulsion which are not included in the RPA expression for the energy width. Let us first consider the exchange correction to the RPA calculation. The lowest-order contribution to the imaginary part of the self-energy is diagrammatically shown in Fig. 7(a); it consists of the direct diagram and the exchange counterpart. As is well known, the self-energy correction corresponding to the direct

diagram is divergent owing to the long-range part of the Coulomb interaction; it can be removed by inclusion of the screening effect. On the other hand, its contribution coming from the short-range part of the interaction is canceled by a half from the corresponding contribution of the exchange counterpart, which is quite similar to the situation in second-order calculation of the ground-state energy. One of the expressions for the energy width which include the exchange contributions as well as the screening effect is written as [see Fig. 7(b) and (c)]

$$\Gamma(p)=\int \frac{d\vec{q}}{(2\pi)^3}[v(q)-\tfrac{1}{2}\langle v(\vec{q}+\vec{p}-\vec{p}\,')\rangle]_{\vec{p}\,\vec{p}\,'}\,\mathrm{Im}\left[\frac{1}{\epsilon_H(q,\epsilon_p-\epsilon_{\vec{p}-\vec{q}})}\right][\Theta(\epsilon_p-\epsilon_{\vec{p}-\vec{q}})-\Theta(\epsilon_F-\epsilon_{\vec{p}-\vec{q}})]$$

$$=\int \frac{d\vec{q}}{(2\pi)^3}v(q)[v(q)-\tfrac{1}{2}\langle v(\vec{q}+\vec{p}-\vec{p}\,')\rangle_{\vec{p}\,\vec{p}\,'}] \tag{32}$$

$$\times\frac{\mathrm{Im}\pi^{(0)}(q,\omega)}{\{1+[v(q)+\tilde{I}_1(q)]\mathrm{Re}\pi^{(0)}(q,\omega)\}^2+\{[v(q)+\tilde{I}_1(q)]\mathrm{Im}\pi^{(0)}(q,\omega)\}^2}$$

$$\times[\Theta(\epsilon_p-\epsilon_{\vec{p}-\vec{q}})-\Theta(\epsilon_F-\epsilon_{\vec{p}-\vec{q}})]\,. \tag{33}$$

The dielectric function $\epsilon_H(q,\omega)$ in Eq. (32) is that of Hubbard,

$$\epsilon_H(q,\omega)=1+v(q)\frac{\pi^{(0)}(q,\omega)}{1+\tilde{I}_1(q)\pi^{(0)}(q,\omega)}\,, \tag{34}$$

$$\tilde{I}_1(q)=\tfrac{1}{2}\langle v(\vec{q}+\vec{p}-\vec{p}\,')\rangle_{\vec{p}\,\vec{p}\,'}\,, \tag{35}$$

where $\pi^{(0)}(q,\omega)$ is the free polarization function. Next, we shall consider the local-field correction arising from the Coulomb repulsion. It can be accomplished by replacing Hubbard's correction

$$\tilde{I}_1(q)=\tfrac{1}{2}\langle v(\vec{q}+\vec{p}-\vec{p}\,')\rangle_{\vec{p}\,\vec{p}\,'}$$

in Eq. (33) by $\tilde{I}(q)$ defined by Eq. (31), so far as one is concerned with the modification of the dielectric function. There is also the other effect of the Coulomb repulsion, which must be treated carefully. Let us look at the factor $v(q)[v(q)-\frac{1}{2}\langle v(\vec{q}+\vec{p}-\vec{p}\,')\rangle_{\vec{p}\,\vec{p}\,'}]$ in Eq. (33). The effect of the Coulomb repulsion is allowed for by replacing every Coulomb interaction in it by the particle-particle ladder vertex [see Fig. 7(d)]. The factor is modified as

$$\langle I(\vec{p},\vec{p}\,';\vec{q})\rangle_{\vec{p}\,\vec{p}\,'}[\langle(\vec{p},\vec{p}\,';\vec{q})\rangle_{\vec{p}\,\vec{p}\,'}-\tfrac{1}{2}\langle I(\vec{p},\vec{p}\,';\vec{q}+\vec{p}-\vec{p}\,')\rangle_{\vec{p}\,\vec{p}\,'}]\,,$$

which cannot be written using only the spin-averaged correction $G(q)$; another correction $C(q)$ arising from spin-antiparallel correlation alone is needed. The resulting expression for Γ is written as

$$\Gamma(p)=\int \frac{d\vec{q}}{(2\pi)^3}v(q)[1-G(q)][1-C(q)]\mathrm{Im}\left[\frac{1}{\epsilon(q,\epsilon_p-\epsilon_{\vec{p}-\vec{q}})}\right][\Theta(\epsilon_p-\epsilon_{\vec{p}-\vec{q}})-\Theta(\epsilon_F-\epsilon_{\vec{p}-\vec{q}})]\,, \tag{36}$$

$$-C(q)v(q)=\langle I(\vec{p},\vec{p}\,';\vec{q})-v(q)\rangle_{\vec{p}\,\vec{p}\,'}\,, \tag{37}$$

$$\epsilon(q,\omega)=1+v(q)\frac{\pi^{(0)}(q,\omega)}{1-G(q)v(q)\pi^{(0)}(q,\omega)}\,. \tag{38}$$

We shall remark here upon the following fact. When the magnitude of $\Gamma(p)$ is not small, the spectral shape of $\mathrm{Im}\tilde{\pi}^{(0)}(q,\omega)$ may be appreciably different from that of $\mathrm{Im}\pi^{(0)}(q,\omega)$. On the other hand, its integrated value over all frequencies does not depend on the magnitude of $\Gamma(p)$ at all. Thus, the energy width of the quasiparticle may produce a significant influence on the spectral shape of $S(q,\omega)$ without changing the static structure factor $S(q)$ appreciably. Finally the following fact should be noted. The quasi-one-pair excitation approximation does not precisely guarantee the re-

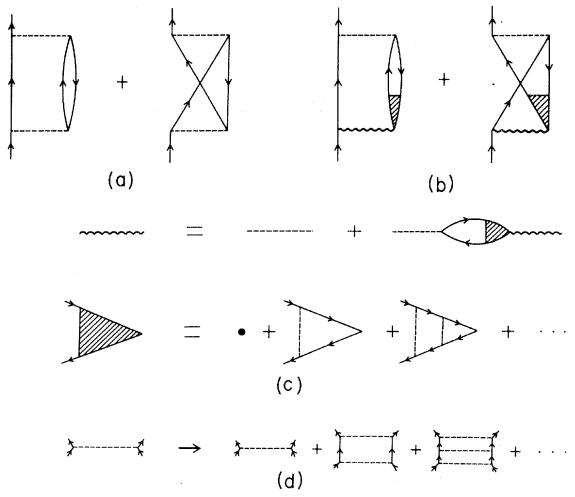


FIG. 7. (a) Lowest-order direct and exchange diagrams contributing to the imaginary part of the self-energy correction. (b) Higher-order self-energy diagrams which include the vertex correction as well as the screened interaction. The wavy line denotes the screened interaction and the shaded triangle the vertex correction. The corresponding exchange counterpart is also shown. (c) The integral equation for the screened interaction is shown diagrammatically. The vertex correction consisting of particle-hole interactions is also shown. (d) The replacement of the bare Coulomb interaction by the particle-particle ladder interaction.

quirement of $g(r) \geq 0$ in the low metallic region, since it does not allow for those types of two-pair excitation processes which have been explained in Sec. II. On the other hand, two-pair excitations and one-pair—one-plasmon excitations arising from the self-energy correction are partially included in a coherent part of the renormalized Green's function; they make a contribution to the energy width of the quasiparticle.

VI. CONCLUDING REMARKS

In this paper we have first discussed critically several sum rules and requirements such as $g(r) \geq 0$, the compressibility sum rule, the frequency moment sum rules, and the conservation law of the local electron number for the purpose of examining whether they are useful practically in developing a dynamical theory of electron correlations. We have next examined in some detail the proper polarization function within the Hartree-Fock approximation. With the intention of having an insight into dynamical aspects of electron correlations, we have investigated lowest-order perturbation terms giving two-pair excitations and one-pair—one-plasmon excitations. It has been pointed out that the renormalization of the one-particle Green's function is indispensable for describing the spectral shape of the dynamical structure factor $S(q,\omega)$ in the intermediate wave-number and intermediate frequency regions. From these considerations we have presented the quasi-one-pair excitation approximation which is expected to be suitable for describing the spectral shape of $S(q,\omega)$ in such a region. In the quasi-one-pair excitation approximation the expression for the proper polarization function is constructed from a coherent part of the renormalized Green's function and a static part of the irreducible particle-hole interaction; internal cancellation between vertex and self-energy correction is taken into account there.

The quasi-one-pair excitation approximation includes effects of two-pair and one-pair—one-plasmon excitations through the energy width of the quasiparticle. Such a coherent contribution from higher-order excitations under the influence of strong short-range correlations represented by the irreducible particle-hole interaction is expected to be essential for the description of the spectral shape of $S(q,\omega)$. The local-field correction $G(q)$ or the irreducible particle-hole interaction and the energy width of the quasiparticle $\Gamma(p)$ are determined in a consistent manner; strong short-range correlations at metallic densities are systematically taken into account in estimating them. The quasi-one-pair excitation approximation has thus been attained as a result of adequate considerations and discussions. A similar form of the proper polarization function has first been proposed by Mukhopadhyay, Kalia, and Singwi,[11] using their own forms of $G(q)$ and $\Gamma(p)$. Their dielectric function is a rather unnatural modification of Vashishta and Singwi's dielectric function which has originally been derived on the basis of an ansatz concerning the two-particle distribution function for the classical system. Physical implication of their modified form of dielectric function is difficult to understand.

In the following paper a numerical study based on the quasi-one-pair excitation approximation is made. The double-peak structure in $S(q,\omega)$ in the intermediate wave-number region as well as the anomalous plasmon dispersion around q_c will be reproduced there in a quite satisfactory manner.

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