Van Hove correlation functions in an interacting electron gas: Equation-of-motion approach

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An extension of the classical van Hove correlation functions to a three-dimensional system of identical fermions is investigated, taking into account interaction effects. This is done within the framework of a Singwi-Tosi-Land-Sjölander-like static local-field approximation, combined with second-order effects of plasmon damping. As a main result the relaxation of the Fermi hole around an instantaneously removed electron is presented.

I. INTRODUCTION

A generalization of the classical Van Hove correlation functions, the *self* and *distinct* parts of the dynamic structure factor, has been investigated recently by Macke *et al.*,[1] concentrating on the free jellium system. A first attempt to calculate interaction effects in the quantum-mechanical self part has been made by Schinner,[2] using (renormalized) first-order perturbation theory. The subject of the present work is the development of a static local-field approximation for the Van Hove functions in the interacting jellium system.

This paper is organized as follows: In Sec. II we write down the basic definitions and the fundamental quantities of our approximation. Using an equation of motion approach leads us to a formula of similar structure as the well-known Singwi-Tosi-Land-Sjölander (STLS) result for the dynamic susceptibility.

An additional factorization step, however, is necessary to evaluate this formula in our case. This will be carried out in Sec. III. As an important result we find an interesting relationship between the short-wavelength limit of our approximation and the classical convolution approximation by Vineyard.[3]

Plasmon damping is built into our model in Sec. IV, using the (renormalized) second-order calculations by Bachlechner *et al.*[4,5] As will be seen from the numerical results presented in Sec. V, the effects of a finite plasmon half-width govern the large-time behavior of the "distinct" correlations. A brief discussion in Sec. VI ends this paper. The following conventions are used.

Energies and momenta are measured in units of $2\epsilon_F$ (ϵ_F denotes the Fermi energy) and k_F (the Fermi momentum), respectively. Excitation spectra (structure factors) are given in units of $mk_F/(2\pi n)$ (m denotes the electron mass, n the mean density). For reasons of brevity spin indices are not written down explicitly. All numerical calculations have been carried out at $r_s=2$, with r_s being the usual density parameter. $\epsilon_{\mathbf{k}}$ and $n_{\mathbf{k}}$ denote the free single-particle excitation energy and the free momentum distribution, respectively, v_q is the Fourier transform of the Coulomb potential. Furthermore, we use the abbreviations $\Delta E_{\mathbf{k},\mathbf{q}}\equiv\epsilon_{\mathbf{k}+\mathbf{q}}-\epsilon_{\mathbf{k}}$ and $\Delta N_{\mathbf{k},\mathbf{q}}\equiv n_{\mathbf{k}}-n_{\mathbf{k}+\mathbf{q}}$. The Lindhard polarizability[6] is denoted by $\chi^0(q,\omega)$.

II. BASIC RELATIONS

A. Definitions

Following Macke *et al.*[1] we write down the decomposition of the dynamic structure factor $S(q,t)$ into its *self* and *distinct* parts $S^s(q,t)$ and $S^d(q,t)$, respectively (for particle number $N\gg 1$),

$$S(q,t)=\frac{1}{N}\sum_{\mathbf{k}}\langle\rho_{\mathbf{q}}(t)c^{\dagger}_{\mathbf{k}}c_{\mathbf{k}-\mathbf{q}}\rangle-N\delta_{\mathbf{q},0}\ , \tag{1}$$

$$S^d(q,t)=\frac{1}{N}\sum_{\mathbf{k}}\langle c^{\dagger}_{\mathbf{k}}\rho_{\mathbf{q}}(t)c_{\mathbf{k}-\mathbf{q}}\rangle-N\delta_{\mathbf{q},0}\ , \tag{2}$$

$$S^s(q,t)=S(q,t)-S^d(q,t)\ , \tag{3}$$

where

$$\rho_q(t)\equiv\sum_{\mathbf{k}}c^{\dagger}_{\mathbf{k},t}c_{\mathbf{k}+\mathbf{q},t} \tag{4}$$

denotes the density operator in field quantization.

For the pair-interaction free-electron gas it can be shown easily that the above definitions of the quantum-mechanical *Van Hove correlation functions* are identical to those valid in the classical system[7,8]

$$S^s(r,t)=\frac{1}{N}\left\langle\sum_i\delta[\mathbf{r}_i(t)-\mathbf{r}_i-\mathbf{r}]\right\rangle\ , \tag{5a}$$

$$S^d(r,t)=\frac{1}{N}\left\langle\sum_{i\neq j}\delta[\mathbf{r}_i(t)-\mathbf{r}_j-\mathbf{r}]\right\rangle-n\ . \tag{5b}$$

There is, however, some evidence that this identity does not hold within an interacting electron gas: The basic concept of the distinct part as the probability of finding two *necessarily different* particles at given locations in space and time is not consistent with quantum physics unless a particle is removed from the system. The definition (5b), however, describes a space-time distribution in equilibrium so that the equivalence with Eq. (2) is questionable in the case when the interaction builds up nontrivial correlations. Consequently, we chose the definition of S^d in field quantization as our starting point.

Although many interesting properties of these functions have been presented for the free fermion system,[1] much less is known about the influence of the interaction.

The renormalized first-order calculations by Schinner[2] provided a first step in this direction: Interaction effects turned out to significantly change the qualitative behavior of the self-correlation function in q-ω space. This theory, however, does not include the plasmon excitation and suffered from a mathematical breakdown in the small-q regime. Thus, the need for a more refined approximation method became obvious.

B. Local-field approximation

Static local-field approximations provide a reliable basic description for static as well as dynamic quantities of the interacting electron gas.[9–13] Although it is well known how such a theory has to be constructed for the density-density response function $\chi(q,\omega)$, it is not obviously seen how this can be accomplished for S^s or S^d. Since both Van Hove functions together contain more information than the dynamic structure factor alone, it is not possible to trivially derive them from, e.g., $\chi(q,\omega)$.

The basic idea of our approximation is to introduce a retarded and an advanced response function,

$$i\sigma^d(q,t)\equiv\theta(t)S^d(q,t)\ , \tag{6a}$$

$$i\tilde{\sigma}^{\,d}(q,t)\equiv-\theta(-t)S^d(q,t)\ , \tag{6b}$$

as counterparts to $\chi(q,\omega)$. Fourier transform of Eq. (6) leads to

$$\overset{(\sim)}{\sigma}{}^d(q,\omega)=\lim_{\eta\to0^+}\frac{1}{2\pi}\int_{-\infty}^{\infty}d\omega'\frac{S^d(q,\omega')}{\omega-\omega'\,{}_{(-)}^{+}i\eta} \tag{7}$$

and, consequently,

$$S^d(q,\omega)=i\sigma^d(q,\omega)-i\tilde{\sigma}^{\,d}(q,\omega)\ . \tag{8}$$

Later on in this section we will restrict our investigations to the case of $S^d(q,\omega)$ having a vanishing imaginary part, so that Eq. (8) reduces to

$$S^d(q,\omega)=-2\,\mathrm{Im}\sigma^d(q,\omega)\ . \tag{9}$$

Equation (9) then clearly shows the close analogy between $\sigma^d(q,\omega)$ and $\chi(q,\omega)$.

As a next step we investigate the equations of motion for σ^d and $\tilde{\sigma}^{\,d}$, respectively. Leaving more detailed calculations to Appendix A we present some important points here: Taking the second time derivative of Eq. (6), Fourier transform and "STLS"-like[14] decoupling lead to

$$\sum_{\mathbf{k}}\Delta E_{\mathbf{k},\mathbf{q}}\left\{(\omega-\Delta E_{\mathbf{k},\mathbf{q}})i\overset{(\sim)}{\sigma}{}^d_{\mathbf{k}}(q,\omega)-i\not{f}_{\mathbf{k},\mathbf{q}}\right.$$
$$\left.-(1-\mathcal{G}_q)v_q\Delta N_{\mathbf{k},\mathbf{q}}\sum_{\mathbf{k}'}i\overset{(\sim)}{\sigma}{}^d_{\mathbf{k}'}(q,\omega)\right\}=0\ , \tag{10a}$$

with

$$i\sigma^d_{\mathbf{k}}(q,t)\equiv\theta(t)\frac{1}{N}\sum_{\mathbf{p}}\langle c^\dagger_{\mathbf{p}}c^\dagger_{\mathbf{k}+\mathbf{q},t}c_{\mathbf{p}-\mathbf{q}}\rangle\ , \tag{10b}$$

$$i\tilde{\sigma}^{\,d}_{\mathbf{k}}(q,t)\equiv-\theta(-t)\frac{1}{N}\sum_{\mathbf{p}}\langle c^\dagger_{\mathbf{p}}c^\dagger_{\mathbf{k}+\mathbf{q},t}c_{\mathbf{p}-\mathbf{q}}\rangle \tag{10c}$$

and

$$\not{f}_{\mathbf{k},\mathbf{q}}\equiv\frac{1}{N}\sum_{\mathbf{p}}\langle c^\dagger_{\mathbf{p}}c^\dagger_{\mathbf{k}}c_{\mathbf{k}+\mathbf{q}}c_{\mathbf{p}-\mathbf{q}}\rangle\ . \tag{10d}$$

$\mathcal{G}_q$ is the STLS static local field

$$\mathcal{G}_q\equiv-\frac{k_F^3}{nv_{\mathbf{q}}q^2}\int\frac{d^3p}{(2\pi)^2}(\mathbf{p}\cdot\mathbf{q})v_{\mathbf{p}}(S_{\mathbf{p}-\mathbf{q}}-1)\ , \tag{10e}$$

$S_{\mathbf{q}}$ denotes the static structure factor.

From its physical meaning the term proportional to $v_{\mathbf{q}}$ in Eq. (10a) is a renormalized correction for each isolated free single-particle excitation with initial momentum $\mathbf{k}$. Since the momentum coupling between the effective single-particle excitations within this approximation is as a whole taken into account by the local field only, the curly bracket in Eq. (10a) must vanish for each $\mathbf{k}$. This immediately leads to

$$\overset{(\sim)}{\sigma}{}^d(q,\omega)=\frac{\overset{(\sim)}{\sigma}{}^{d0}_V(q,\omega)}{1-(1-\mathcal{G}_q)v_q\overset{(\sim)}{\chi}{}^0(q,\omega)} \tag{11}$$

with

$$\overset{(\sim)}{\sigma}{}^{d0}_V(q,\omega)\equiv\lim_{\eta\to0^+}\sum_{\mathbf{k}}\frac{\not{f}_{\mathbf{k},\mathbf{q}}-n_{\mathbf{k}}\delta_{\mathbf{q},0}}{\omega-\Delta E_{\mathbf{k},\mathbf{q}^+_{(-)}}i\eta} \tag{12a}$$

and

$$\tilde{\chi}^0(q,\omega)=[\chi^0(q,\omega)]^*\ . \tag{12b}$$

Of course, the more comprehensive way of decoupling the equations of motion for the Wigner matrix[15] $\rho_{\mathbf{k},\mathbf{q}}(t)\equiv c^\dagger_{\mathbf{k},t}c_{\mathbf{k}+\mathbf{q},t}$, instead of those for the density $\rho_{\mathbf{q}}(t)$, would have led to the same result Eqs. (11) and (12).

Again we note the close resemblance between Eq. (11) and the well-known result for $\chi(q,\omega)$

$$\chi(q,\omega)\simeq\frac{\chi^0(q,\omega)}{1-(1-\mathcal{G}_q)v_q\chi^0(q,\omega)}\ . \tag{13}$$

Contrary to Eq. (13) we, however, *cannot* simply approximate $\sigma^{d0}_V(q,\omega)$ in Eq. (11) by its interaction-free value: As will be seen later this would lead to an unsatisfactory result for the static limit of S^d. Since any static local-field approximation neglects energy- and momentum-coupled multipair processes, it is consistent with this to neglect such processes in S^{d0}_v too. Consequently, the expectation value $\not{f}_{\mathbf{k},\mathbf{q}}$ in Eq. (12a) will have a vanishing imaginary part, which immediately leads to

$$\tilde{\sigma}^{\,d0}_V(q,\omega)=[\sigma^{d0}_V(q,\omega)]^* \tag{14}$$

and the validity of Eq. (9). Furthermore, the real and imaginary parts of $\sigma^{d0}_V(q,\omega)$ obey the Kramers-Kronig relation

$$\mathrm{Re}\sigma^{d0}_V(q,\omega)=\frac{1}{2\pi}\,\mathrm{P}\int_{-\infty}^{\infty}d\omega'\frac{S^{d0}_V(q,\omega')}{\omega-\omega'}\ , \tag{15}$$

where $S^{d0}_V(q,\omega)=-2\,\mathrm{Im}\sigma^{d0}_V(q,\omega)$.

C. Further investigations

Combining Eqs. (9) and (11) we find

$$S^d(q,\omega)=S_V^{d0}(q,\omega)\frac{e_1(q,\omega)}{e_1(q,\omega)^2+e_2(q,\omega)^2}$$

$$+2\,\mathrm{Re}\sigma_V^{d0}\frac{e_2(q,\omega)}{e_1(q,\omega)^2+e_2(q,\omega)^2} \tag{16}$$

with

$$e(q,\omega)\equiv 1-v_q(1-\mathcal{G}_q)\chi^0(q,\omega)\ , \tag{17a}$$

$$e_1(q,\omega)\equiv \mathrm{Re}e(q,\omega),\quad \text{and } e_2(q,\omega)\equiv \mathrm{Im}e(q,\omega)\ . \tag{17b}$$

Obviously, the quantity $e(q,\omega)$ as introduced above is connected with the dielectric function $\epsilon(q,\omega)$ by the relation

$$\epsilon(q,\omega)=e(q,\omega)\frac{1-\mathcal{G}_q}{1-\mathcal{G}_q e(q,\omega)}\ , \tag{18}$$

and is identical with $\epsilon(q,\omega)$ in the case of vanishing local-field (random-phase approximation, RPA).

Leaving the detailed discussion of how to construct a realistic approximation for $\sigma_V^{d0}(q,\omega)$ to the next section, we obtain the following results by a closer inspection of Eq. (16).

(i) The most important quantities involved in our theory have the symmetry properties: (a) $S^d(q,\omega)$, $e_1(q,\omega)$, and $\mathrm{Im}\sigma_V^{d0}(q,\omega)$ are *even* functions in ω; (b) $e_2(q,\omega)$ and $\mathrm{Re}\sigma_V^{d0}(q,\omega)$ are *odd* functions in ω. Consequently, $S^d(q,t)$ is real and even with respect to t. From the analysis by Macke *et al.*[1] it is known that within second-order perturbation theory this symmetry is broken, i.e., $S^d(q,-t)\neq S^d(q,t)$. The physical meaning of this is still not known and will be left to future investigations, as including that type of multipair process lies beyond the scope of the present work.

(ii) The term proportional to e_2 in Eq. (16) describes the plasma excitation (plasmon). Its dispersion relation is the same as in the corresponding theories for $S(q,\omega)$. Due to the symmetry properties (i) there are, however, *two* plasmon peaks at frequencies $\omega=\pm\omega_{\mathrm{pl}}(q)$, which are undamped outside the particle-hole continuum. This behavior is especially unsatisfactory, as we want to describe the (nonlinear) dynamic response of the system after one electron has been removed instantaneously: The undamped plasma oscillations dominate the space-time response function on an unphysical long time scale. Consequently, in Sec. IV we shall build energy- and momentum-coupled second-order processes into our theory.

From Eq. (16) we get the plasmon strength $Z^d(q)$,

$$Z^d(q)=2\pi\,\mathrm{Re}\sigma_V^{d0}[q,\omega_{\mathrm{pl}}(q)]$$
$$\times\left[\frac{\partial}{\partial\omega}e_1(q,\omega)\bigg|_{\omega=\omega_{\mathrm{pl}}(q)}\right]^{-1}\theta(q_c-q)\ , \tag{19}$$

where q_c is the plasmon critical wave vector. In the $q\to 0$ limit Eq. (19) can be evaluated without detailed knowledge about σ_V^{d0}, leading to $Z^d(q\to 0^+)=-\frac{2}{3}$. Contrary to the plasma excitation in $S(q,\omega)$ the plasmon strength in $S^d(q,\omega)$ does not vanish for finite, but arbitrarily small q. As a consequence we find

$$S^d(q,t)=-\cos(\omega_p t)+O(q^2)\ , \tag{20}$$

with ω_p being the classical plasmon frequency. This result seems to stand in contradiction to the exact normalization condition $S^d(q=0,t)=-1$. A more detailed mathematical analysis, however, shows[16] that Eq. (20) is valid even within the exact electron gas. The $q\to 0$ limit does not exist in the mathematical sense which means that, despite the validity of Eq. (20), the normalization requirement is indeed fulfilled at the isolated point $q=0$. This pathology originates from the fact that the $q=0$ contribution of the two-particle interaction v_q in the jellium system is exactly canceled by the background charge.

III. FACTORIZATION APPROACH FOR σ_V^{d0}

In the well-studied local-field approximation[9] for the dynamic susceptibility $\chi(q,\omega)$ the numerator (i.e., the inhomogeneity of the equation of motion) is usually evaluated within the free system [cf. Eq. (13)]. Although it is known that this procedure causes some problems concerning the third moment sum rule,[17] the major aspects of χ are nevertheless well described.

In the case of our theory, however, one finds from Eq. (11) that the inhomogeneity σ_V^{d0} *alone* determines the static limit, i.e.,

$$S^d(q,t=0)=S_q-1=S_V^{d0}(q,t=0)\ , \tag{21}$$

the proof of which will be given in Appendix B. Therefore it is clear that evaluating σ_V^{d0} just within the interaction-free system we would impose an unphysical initial condition on the behavior of our time-dependent response functions. Again, Eq. (21) shows that the task of realistically approximating this quantity is a highly nontrivial one, since comprehensive calculations are necessary in order to obtain a good static structure factor S_q.[9-13]

Consequently we will use S_q as an input into our theory, obtained from the best available results, namely the Monte Carlo calculations by Ceperley and Alder.[18-20] Since the quantity σ_V^{d0} is a combination of an "exact" expectation value with a free system's time dependence, the following factorization is suggested:

$$S_V^{d0}(q,\omega)\simeq\Gamma_q S^{d0}(q,\omega)$$
$$-(1-\Gamma_q)\tfrac{1}{2}[S^{s0}(q,\omega)+S^{s0}(q,-\omega)]\ . \tag{22}$$

Herein S^{d0} and S^{s0} denote the free distinct and self-parts, respectively. Γ_q is chosen according to Eq. (21),

$$\Gamma_q = \frac{S_q}{S_q^0} , \tag{23}$$

S_q^0 being the free static structure factor.

The physical meaning of the ansatz Eq. (22) becomes more evident when rewriting the quantity $S_V^{d0}(q,\omega)$ as

$$S_V^{d0}(q,\omega) \simeq -\frac{2\pi}{N} \sum_{\mathbf{k}} \mathcal{W}_{\mathbf{k},\mathbf{k}+\mathbf{q}} \delta(\omega - \Delta E_{\mathbf{k},\mathbf{q}}) , \tag{24}$$

with

$$\mathcal{W}_{\mathbf{k}_1,\mathbf{k}_2} \simeq \begin{cases} 1 \text{ for } k_1 < 1 \text{ and } k_2 < 1 \\ \frac{1}{2}(1 - \Gamma_{|\mathbf{k}_2 - \mathbf{k}_1|}) \text{ for } k_1 < 1 \text{ and } k_2 > 1 \text{ (or } k_2 < 1 \text{ and } k_1 > 1) \\ 0 \text{ otherwise} . \end{cases} \tag{25}$$

$\mathcal{W}_{\mathbf{k}_1,\mathbf{k}_2}$ then is the "probability" that an excitation from momentum $\mathbf{k}_1$ to momentum $\mathbf{k}_2$ is *forbidden*. In the free system ($\Gamma_q = 1$) only the excitation process between two occupied momentum levels is forbidden. Interaction effects are taken into account by reducing the probability for an excitation to occur even in the case when a filled and an empty Fermi sphere overlap.

Although this interpretation gives strong evidence for the physical meaningfulness of the ansatz Eq. (22), a quantitative analysis is still pending. As a systematic approach we evaluate Eqs. (10d) and (12a) within (renormalized) first-order perturbation theory. This method of substituting an effective (screened) static potential into the expressions of finite order perturbation theory has already been applied successfully in various fields, e.g., the first-order analysis by Schinner,[2] and the plasmon damping calculations by Bachlechner, Böhm, and Schinner.[21]

Leaving the detailed calculations to Appendix C we present only the results here: In Fig. 1 we compare various approximations for $S_V^{d0}(q,\omega)$, namely the first-order perturbation theory with bare Coulomb potential and screened Thomas-Fermi potential, respectively, as well as the free system and the ansatz Eq. (22). We note a good conformity of the factorization approach with the results of the renormalized first-order theory. As expected, the unscreened first-order calculations show the well-known breakdown in the region of small q (cf. Ref. 2).

Although both theories, the renormalized first-order analysis and Eq. (22), provide good descriptions of the quantity S_V^{d0} in Fourier space, the former has a great disadvantage as we look at the real-space correlations: Not even the use of a screened potential can prevent the pair-correlation function at zero particle separation $g(0)$ from becoming negative at metallic densities, when evaluated within first-order perturbation theory. Consequently we shall use Eq. (22) within the following calculations.

Finally, we want to point out another very interesting consequence of our ansatz Eq. (22): Noting that for $q > 2$ the free distinct part $S^{d0}(q,\omega)$ exactly vanishes, and that the free static structure factor equals one, we immediately find

$$S_V^{d0}(q,t) = (S_q - 1)\mathrm{Re}S^{s0}(q,t) \quad \text{for} \quad q > 2 . \tag{26}$$

This relation can be seen as a quantum-mechanical counterpart to the very successful classical convolution approximation by Vineyard.[3] Equation (26) also holds for the total $S^d(q \gg 2,t)$ within our approximation, as can be derived directly from Eq. (16). It therefore reflects the classical limit of our model. Further investigations are currently in progress to show whether these *quasiclassical correlations* in the Fermi fluid can lead to new types of approximations for other quantities of the quantum-mechanical many-body system, too.

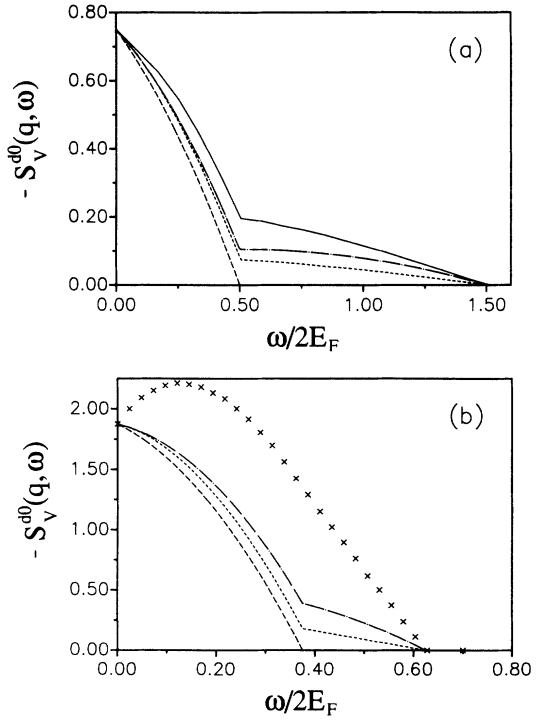

FIG. 1. Plot of the function S_V^{d0} vs ω for fixed q. The curves correspond to bare first-order theory (full line and crosses, respectively), screened first-order theory (short-dashed line), the factorization ansatz Eq. (22) (dash-dotted line), and the free system (long-dashed line). (a) $q = 1$, (b) $q = 0.5$.

IV. PLASMON DAMPING

As we have mentioned earlier, the scope of the present paper is to evaluate the quantum-mechanical Van Hove functions for the interacting Fermi system within a static local-field approximation. This type of approach of course neglects energy- and momentum-coupled multipair excitations, leading, for example, to plasmon damping and a broken time symmetry of $S^d(r,t)$. Leaving the latter to future investigations we concentrate here on building a finite plasmon lifetime into our theory, since we believe this effect to be of special physical relevance.

Recent progress has been made in understanding plasmon decay in simple metals, both theoretical[22–24] and experimental.[25–27] Following the work of Bachlechner *et al.*[5] we write down the plasmon full width at half maximum $\Delta E_{1/2}$ for an electron gas

$$\Delta E_{1/2} \cong \omega_p \mathrm{Im}\ \epsilon(q,\omega_p) \equiv \omega_p b q^2 + O(q^4) \ . \tag{27}$$

The main contribution to the q^2 coefficient b arises from the decay of the plasmon into two electron-hole pairs. It can be investigated within the formalism of second-order perturbation theory and yields[21]

$$\mathrm{Im}\ \epsilon^{2\ \mathrm{pairs}}(q,\omega_p) \simeq \frac{9\omega_p^2 q^2}{5\pi 2^5} \int_0^\infty dp \int d^3k \int d^3k' n_{\mathbf{k}} n_{\bar{\mathbf{k}}-\mathbf{p}} n_{\mathbf{k}'} n_{\bar{\mathbf{k}}'+\mathbf{p}} \times \delta(\omega_p + \mathbf{p}'\cdot\mathbf{p}) p^2 \mathcal{B} \ , \tag{28}$$

where

$$\mathcal{B} = (v_p^{\mathrm{se}})^2 \mathcal{B}^{\mathrm{se}} - \tfrac{1}{2} v_p^{\mathrm{ex}} v_{p'}^{\mathrm{ex}} \mathcal{B}^{\mathrm{ex}} \ , \tag{29a}$$

$$\mathcal{B}^{\mathrm{se}} = \tfrac{7}{2} - a_p^{\mathrm{se}} + \tfrac{3}{2}(a_p^{\mathrm{se}})^2 + \frac{4}{\omega_p^2}[k^2p^2 - (\mathbf{k}\cdot\mathbf{p})^2] \ , \tag{29b}$$

$$\mathcal{B}^{\mathrm{ex}} = \tfrac{3}{2} - a_p^{\mathrm{ex}} + \tfrac{1}{2} a_p^{\mathrm{ex}} a_{p'}^{\mathrm{ex}} + \frac{\omega_p^2}{p^2 p'^2} a_p^{\mathrm{ex}} a_{p'}^{\mathrm{ex}} + 2\frac{p^2 p'^2}{\omega_p^2} \ , \tag{29c}$$

$$a_p^{\mathrm{se/ex}} \equiv \frac{p\frac{\partial}{\partial p} v_p^{\mathrm{se/ex}}}{v_p^{\mathrm{se/ex}}} \tag{29d}$$

for the imaginary part of the dielectric function $\epsilon(q,\omega_p)$. In Eqs. (28) and (29) a potential renormalization step similar to that of the first-order calculations in Sec. III and Ref. 2 has been carried out. This means that the bare Coulomb interaction has been replaced by a static, but spin-dependent, potential $v_p^{\mathrm{se/ex}}$ for the direct and exchange contributions, respectively. Using the effective potential derived by Singwi[28] then leads to a good agreement of this theory with the experiment.[21]

Now, to build these results into our model we note that the plasmon peaks in $S^d(q,\omega)$ are well separated from the contributions of the particle-hole continuum, except when $q \approx q_c$. This allows us to simply replace the δ peaks in the corresponding term of Eq. (16) by

$$S^d_{\mathrm{pl}}(q,\omega) = Z^d(q)\{F_\Gamma[\omega - \omega_{\mathrm{pl}}(q)] + F_\Gamma[\omega + \omega_{\mathrm{pl}}(q)]\} \ , \tag{30}$$

where $F_\Gamma(\omega)$ with $\Delta E_{1/2} = 2\Gamma$ denotes the Lorentzian shape of the plasmon peaks

$$F_\Gamma(\omega) = \frac{1}{\pi}\frac{\Gamma}{\omega^2 + \Gamma^2} \tag{31}$$

and $Z^d(q)$ is given by Eq. (19).

V. NUMERICAL RESULTS

In this section we present various numerical results of our theory. The calculations were carried out by combining Eq. (16) and the factorization ansatz Eq. (22). Therein the static structure factor was obtained from the Monte Carlo results by Ceperley and Alder.[18–20] The local field $\mathcal{G}_q$ was either set to zero (referred to as "RPA") or has been calculated from Eq. (10e) (referred to as "STLS"). Furthermore, the plasmon contributions were separated from Eq. (16) by using Eq. (30) and the damping theory presented in Sec. IV.

(i) In Fig. 2 we compare $S^d(q,\omega)$ for "STLS" and "RPA" local fields with the free system. It is especially noteworthy that the distinct part in the interacting system has a nonvanishing spectral density within the complete particle-hole continuum ($|\omega| \le q + q^2/2$) while $S^{d0}(q,\omega)$ equals zero for $|\omega| \ge q - q^2/2$.

(ii) Calculating the full dynamic structure factor $S(q,\omega)$ within the well-known STLS approximation, Eq. (3) and (i) then immediately yield the self part $S^s(q,\omega)$. It is of great interest to compare this result with the previous analysis in renormalized first-order perturbation theory:[2] Fig. 3 shows the remarkable agreement of both completely different approaches, which we take as addi-

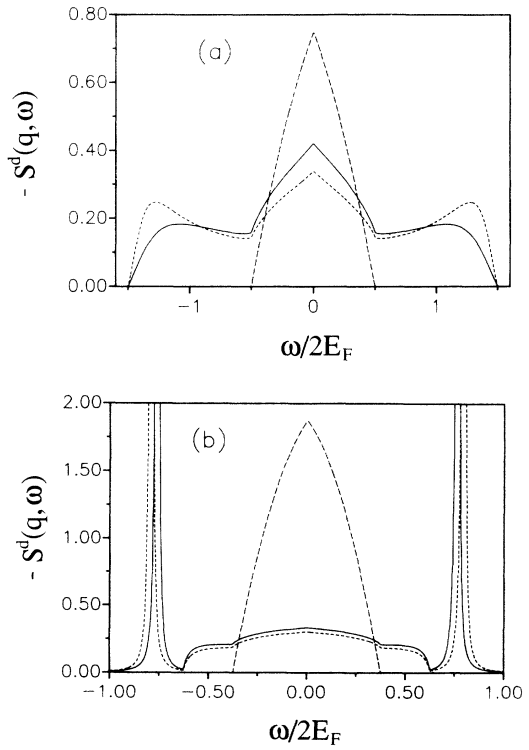

FIG. 2. Plot of the distinct part $S^d(q,\omega)$ vs ω for fixed q. The curves correspond to the local-field approximations (cf. Sec. V) "STLS" (full line), "RPA" (short-dashed line), and the free system (long-dashed line). (a) $q = 1$, (b) $q = 0.5$.

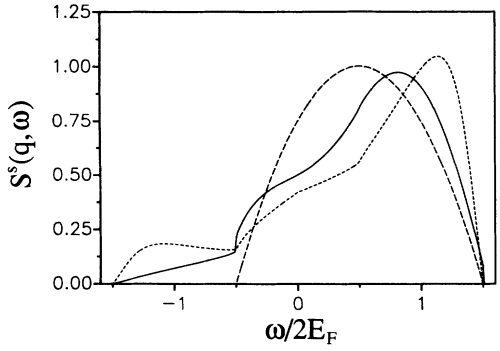

FIG. 3. Comparison of the self-part at $q=1$ for renormalized first-order perturbation theory (Ref. 2) (full line), the present "STLS" approximation (short-dashed line), and the free system (long-dashed line).

tional strong evidence for the validity of our approximations.

(iii) With the use of $S^d(q,t)$ another very interesting quantity can be calculated, i.e., a time-dependent generalization of the static pair-correlation function $g(r)$:

$$g(r,t)\equiv 1+\frac{3}{2}\frac{1}{r}\int_0^\infty dq\, q\,\sin(qr)S^d(q,t)\ . \tag{32}$$

Obviously, $g(r,t=0)$ reduces to its static counterpart $g(r)$. From its physical meaning Eq. (32) describes how the Fermi hole around an electron is filled up when this electron has been removed instantaneously at time $t=0$. In Fig. 4, $g(r,t)$ is plotted versus r for various times.

(iv) This "decay" of the Fermi hole can be studied even better by looking at another quantity closely related to $g(r,t)$:

$$N_R(t)\equiv\frac{4}{3\pi}\int_0^R dr\, r^2[1-g(r,t)]\ . \tag{33}$$

$N_R(t)$ is the total charge (in electron charges) within a sphere of radius R. Setting R to the average spacing between the electrons, i.e., $R=2r_s a_B$ (a_B denotes the Bohr radius), one obtains the results presented in Fig. 5. It is seen that the plasma oscillations dominate the behavior of this quantity. A more detailed analysis shows that the decay of $N_R(t)$ for large times obeys a power law, namely

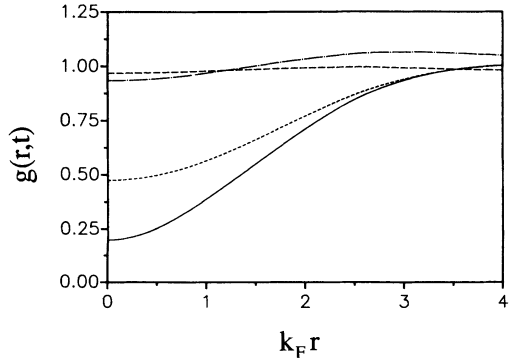

FIG. 4. The generalized pair distribution function $g(r,t)$ vs r for various times: $t=0$ (full line), $t=1$ (short-dashed line), $t=5$ (dash-dotted line), $t=10$ (long-dashed line).

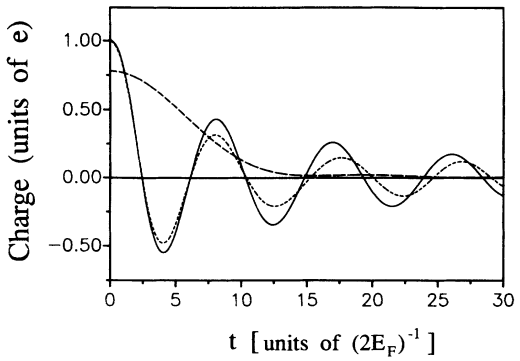

FIG. 5. Total charge (in electron charges) within a sphere of radius $2r_s a_B$ vs time. The curves correspond to the present theory using screened plasmon damping (full line), unscreened damping (short-dashed line), and the free system (long-dashed line).

$$N_R(t)\simeq\frac{R^3}{6\pi^{1/2}}(\gamma t)^{-3/2}\cos(\omega_p t)\ \text{ for }\ \gamma t\gg 1\ , \tag{34}$$

where $\gamma\equiv b\omega_p/2$ with b from Eq. (27). Since for the renormalized model (cf. Sec. IV) one obtains $\gamma\approx 10^{-2}$, the full curve in Fig. 5 would remain almost unchanged when simply setting $\gamma=0$. This just reflects the fact that the plasmon is a well-defined excitation. Nevertheless, the asymptotic behavior (i.e., the envelope curve of the oscillations) *is* of significant physical relevance, especially from the experimentalist's point of view, which proves the great importance of a refined plasmon damping model within our theory.

VI. CONCLUSIONS

The subject of this paper was a closer investigation of the Van Hove correlation functions in an interacting many-fermion system. Contrary to the well-known static pair-correlation function, the physical interpretation and content of its time-dependent counterpart Eq. (32) is not so obvious. Its importance, however, mainly lies in the direct connection of classical liquid-state concepts with quantum systems: The distinct part of the dynamic structure factor as defined by Eq. (2) describes the probability of finding two different electrons separated by given distances in space *and* time. Later on in this section we shall point out some possible experimental applications that can make use of the knowledge of this function. Furthermore, it could turn out to be of even greater interest to use our present results within new dynamic approximations (see, e.g., the work by Kerr;[29] see also the discussion in Sec. VI of Ref. 1). Coarsely spoken, any multiparticle process that involves the quasi-instantaneous removal of a particle is a promising candidate for applying the ideas pointed out within this paper.

The mathematical analysis of these functions was done within the framework of a static local-field approximation. Although this type of approach can be derived straightforwardly for the total excitation probability, i.e., the dynamic structure factor $S(q,\omega)$, in the case of its distinct part unexpected problems arise: The numerator

in the well-known STLS result Eq. (13) has to be replaced by a much more complicated quantity, in order to obtain the counterpart Eq. (11). This function $\sigma_V^{d0}(q,\omega)$ cannot be approximated by its interaction-free value since it contains the initial condition for $t=0$. Furthermore, the long-wavelength limit ($q\to 0$) reflects a fundamental pathology of the jellium system.

To evaluate the function σ_V^{d0} the factorization ansatz Eq. (22) has been made, motivated by a microscopic analysis. As an important consequence of this, in the short-wavelength limit ($q\to\infty$) our theory reduces to a generalization of the classical Vineyard approximation. We call this a *quasiclassical* limit since in Eq. (26) the self-part still has a nonvanishing imaginary part. We believe that further investigations of this type will lead to a deeper understanding of how to generalize classical approximations to the quantum many-body problem.

Despite the fact that our theory gives reasonable results for the principal structure of the investigated functions, some important questions could not yet be answered: Although we were able to build a finite plasmon lifetime into our model, those terms leading to a broken time symmetry had to be neglected within this framework. A dynamic local-field approximation or, better, a Baym-Kadanoff analysis, as done by Green, Neilson, and Szymanski[30] for $S(q,\omega)$, will most probably reveal new aspects of this topic.

Furthermore, it will be of great importance to investigate how the "distinct correlations" can be accessed by the experiment (the self part is complex in the exact system and thus not observable). Obviously, a direct measurement of, e.g., the time-dependent pair-correlation function Eq. (32) is a highly nontrivial task. In this case one most probably will have to rely on computer experiments, as is also necessary already for the usual static pair function. However, it should be possible to find traces of the relaxation process described by Eq. (33) in, e.g., the photoelectric effect or electron-positron annihilation in simple metals.

Perhaps the most promising way to gain experimental access to the quantities discussed here could lie in the field of ion-induced electron emission and secondary-electron emission:[31] In theoretical models for the secondary-electron yield the response of the electron gas to instantaneously removing electrons turns out to be of great relevance.[32] Investigations to build our results into these theories are currently in progress.

It is, however, beyond the scope of the present paper to discuss experimental setups in a more comprehensive way, since our intentions were to concentrate on analyzing special aspects of the internal structure of the electron gas. Consequently, from our point of view the most important result is that the classical concept of *self*-motion and *distinct* motion can be successfully generalized to the case of an interacting quantum fluid.

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APPENDIX A

In this appendix we give a brief description of the major steps leading to Eq. (10). Following the work of Singwi *et al.*[14] we start by writing equations of motion for the density operator:

$$i\dot{\rho}_q(t)=\sum_{\mathbf{k}}\Delta E_{\mathbf{k},\mathbf{q}}c^{\dagger}_{\mathbf{k},t}c_{\mathbf{k}+\mathbf{q},t}\ , \tag{A1}$$

$$-\ddot{\rho}_q=\sum_{\mathbf{k}}(\Delta E_{\mathbf{k},\mathbf{q}})^2c^{\dagger}_{\mathbf{k},t}c_{\mathbf{k}+\mathbf{q},t} +v_q(1-\mathcal{G}_q)\sum_{\mathbf{k}}\Delta E_{\mathbf{k},\mathbf{q}}\Delta N_{\mathbf{k},q}\rho_q(t)\ . \tag{A2}$$

Therein $\mathcal{G}_q$ is given by Eq. (10e). It is especially noteworthy that Eq. (A1) just reflects the particle conservation and, consequently, is exact. To derive Eq. (A2) the terms nonlinear in ρ have been approximated by factorized expectation values.

Taking the second time derivative of the quantity $\pm\theta(\pm t)\{S^d(q,t)-S^d(q,0)\}$ yields

$$i\frac{\partial}{\partial t}\theta(\pm t)\langle c^{\dagger}i\dot{\rho}_q(t)c\rangle =\pm\delta(t)\langle c^{\dagger}i\dot{\rho}_q(0)c\rangle-\theta(\pm t)\langle c^{\dagger}\ddot{\rho}_q(t)c\rangle\ , \tag{A3}$$

where for brevity reasons the momentum dependence of the $c^{\dagger}$ and c operators and the corresponding summations are omitted. Inserting Eqs. (A1) and (A2) into Eq. (A3) then immediately leads to Eq. (10a).

It should be noted that from Eqs. (A1) and (A2), of course, the well-known STLS result for the dynamic susceptibility can also be derived: Starting from the definition of $\chi(q,t)$

$$\chi(q,t)\equiv-i\theta(t)\langle[\rho_q(t),\rho_{-q}(0)]_-\rangle\ , \tag{A4}$$

and again expressing $\dot{\rho}$ and $\ddot{\rho}$ in the second term derivative of Eq. (A4) by Eqs. (A1) and (A2), we arrive at a counterpart of Eq. (10a), from which Eq. (13) is easy to derive.

APPENDIX B

The subject of this appendix is to prove Eq. (21). We start by defining an auxiliary quantity

$$\sigma_{\lambda}^{d}(q,\omega)\equiv\frac{\sigma_V^{d0}(q,\omega)}{1-\lambda v_q(1-\mathcal{G}_q)\chi^0(q,\omega)}\ , \tag{B1}$$

where λ is an arbitrary real number. Now, introducing

$$S_{\lambda}^{d}(q,\omega)\equiv-2\,\mathrm{Im}\sigma_{\lambda}^{d}(q,\omega)\ , \tag{B2}$$

our task reduces to the proof of

$$\frac{\partial}{\partial\lambda}S_{\lambda}^{d}(q,t=0)=0\ , \tag{B3}$$

since from Eq. (B3) the equal sign in Eq. (B4)

$$S^d(q,t=0)\equiv S^d_{\lambda=1}(q,t=0)=S^d_{\lambda=0}(q,t=0)$$
$$\equiv S_V^{d0}(q,t=0) \tag{B4}$$

follows immediately. The left-hand side of Eq. (B3) can be written as

$$\frac{\partial}{\partial\lambda}S^d_\lambda(q,t=0)\propto v_q(1-\mathcal{G}_q)\mathrm{Im}\int_{-\infty}^{\infty}d\omega\,\sigma^d_\lambda(q,\omega)\chi_\lambda(q,\omega)\ , \tag{B5}$$

where we have introduced the quantity

$$\chi_\lambda(q,\omega)\equiv\frac{\chi^0(q,\omega)}{1-\lambda v_q(1-\mathcal{G}_q)\chi^0(q,\omega)} \tag{B6}$$

in analogy to Eq. (B1). Using the Kramers-Kronig relation Eq. (15), that is valid also for χ_λ and σ^d_λ, yields

$$\frac{\partial}{\partial\lambda}S^d_\lambda(q,t=0)\propto\int_{-\infty}^{\infty}d\omega\int_{-\infty}^{\infty}d\omega'\frac{1}{\omega-\omega'}\{\mathrm{Im}\sigma^d_\lambda(\omega)\mathrm{Im}\chi_\lambda(\omega')+\mathrm{Im}\sigma^d_\lambda(\omega')\mathrm{Im}\chi_\lambda(\omega)\}\ . \tag{B7}$$

The right-hand side of Eq. (B7) is identical to zero which is easily seen by exchanging ω and ω'.

APPENDIX C

Our aim is to evaluate the quantity $S_V^{d0}(q,\omega)$ within first-order perturbation theory. As a starting point we write down the spectral theorem[33]

$$\langle A(t)B(0)\rangle=[e^{i\beta\partial/\partial t}+1]^{-1}\langle[A(t),B(0)]_+\rangle\ , \tag{C1}$$

where β denotes the inverse temperature; $A(t)$ and $B(t)$ are operators in the Heisenberg representation. Applying Eq. (C1) to the expectation value $\langle c_1^\dagger c_2\rangle$ and neglecting terms of order v^2 in the anticommutator leads to ($\beta\to\infty$)

$$n_1(\mu)=\theta(\mu-\epsilon_{\mathbf{k}_1})+O(v^2) \tag{C2}$$

for the momentum distribution. μ denotes the chemical potential in first-order theory. A similar calculation yields

$$\langle c_1^\dagger c_2^\dagger c_3 c_4\rangle=n_1n_2(\delta_{14}\delta_{23}-\delta_{13}\delta_{24})+\frac{n_1n_2(n_3-n_4^-)-n_3n_4(n_1-n_2^-)}{\epsilon_4+\epsilon_3-\epsilon_2-\epsilon_1}$$
$$\times\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4}\{v_{\mathbf{k}_4-\mathbf{k}_1}\delta_{\sigma_1\sigma_4}\delta_{\sigma_2\sigma_3}-v_{\mathbf{k}_4-\mathbf{k}_2}\delta_{\sigma_1\sigma_3}\delta_{\sigma_2\sigma_4}\}+O(v^2)\ . \tag{C3}$$

Herein n^- is an abbreviation for $1-n$, and $1\equiv\mathbf{k}_1,\sigma_1$.

Using expression (C3) we finally obtain

$$S_V^{d0}(q,\omega)=S^{d0}(q,\omega)+\Delta S_V^{d0}(q,\omega)+O(V^2) \tag{C4}$$

with [$\alpha\equiv(4/9\pi)^{1/3}$]

$$\Delta S_V^{d0}(q,\omega)\equiv\left[\frac{\alpha r_s}{\pi^3}\right]\frac{2\pi}{q^2}\theta(1+q-|\nu_+|)$$
$$\times\int_0^{2+q}dp\int_{-1}^{1}d\zeta_{pq}\int_{|\nu_+|}^{1+q}dk\int_0^{2\pi}d\varphi\left[v_q-\frac{v_p}{2}\right]$$
$$\times\left[\frac{pk}{\zeta_{pq}}\right]\{n_{\mathbf{k}+\mathbf{q}-\mathbf{p}}n_{\mathbf{k}}(n_{\mathbf{k}+\mathbf{q}}-n^-_{\mathbf{k}-\mathbf{p}})-n_{\mathbf{k}+\mathbf{q}}n_{\mathbf{k}-\mathbf{p}}(n_{\mathbf{k}+\mathbf{q}-\mathbf{p}}-n_{\mathbf{k}}^-)\} \tag{C5}$$

and

$$\mathbf{k}\cdot\mathbf{q}=q\nu_+,\quad\mathbf{k}\cdot\mathbf{p}=p\nu_+\zeta_{pq}+p\sqrt{k^2-\nu_+^2}\sqrt{1-\zeta_{pq}^2}\cos(\varphi),\quad\mathbf{p}\cdot\mathbf{q}=pq\zeta_{pq},\quad\nu_+\equiv\frac{\omega}{q}-\frac{q}{2}\ . \tag{C6}$$

From Eqs. (C4) to (C6) the numerical results of Fig. 1 have been calculated, using a Monte Carlo algorithm to carry out the fourfold integral.

[1]W. Macke, H. M. Miesenböck, K. Hingerl, and M. E. Bachlechner, Phys. Rev. B **39**, 2117 (1989).
[2]A. Schinner, Phys. Rev. B **41**, 2524 (1990); **43**, 1211(E) (1991).
[3]G. H. Vineyard, Phys. Rev. **110**, 999 (1958).
[4]M. E. Bachlechner, Diploma thesis, Johannes Kepler Universität, Linz, 1990.
[5]M. E. Bachlechner, W. Macke, H. M. Miesenböck, and A. Schinner, Physica B **168**, 104 (1991).
[6]J. Lindhard, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. **28**, 5 (1954).
[7]L. van Hove, Phys. Rev. **95**, 249 (1954).
[8]S. W. Lovesey, *Theory of Neutron Scattering from Condensed Matter* (Clarendon, Oxford, 1984).
[9]K. S. Singwi and M. P. Tosi, Solid State Phys. **36**, 177 (1981).
[10]S. Ichimaru, Rev. Mod. Phys. **54**, 1017 (1982).
[11]G. D. Mahan, *Many-Particle Physics* (Plenum, New York, 1990).
[12]N. Iwamoto and D. Pines, Phys. Rev. B **29**, 3924 (1984).
[13]N. Iwamoto, E. Krotschek, and D. Pines, Phys. Rev. B **29**, 3936 (1984).
[14]K. S. Singwi, M. P. Tosi, R. H. Land, and A. Sjölander, Phys. Rev. **176**, 589 (1968).
[15]See, e.g., A. A. Kugler, J. Stat. Phys. **12**, 35 (1975).
[16]A. Schinner, Doctoral thesis, Johannes Kepler Universität, Linz, 1991.
[17]A. Holas, in *Strongly Coupled Plasma Physics,* edited by F. J. Rogers and H. E. DeWitt (Plenum, New York, 1988).
[18]D. Ceperley, Phys. Rev. B **18**, 3126 (1978).
[19]D. Ceperley and B. Alder, Phys. Rev. Lett. **45**, 566 (1980).
[20]D. Ceperley (private communication).
[21]M. E. Bachlechner, H. M. Böhm, and A. Schinner (unpublished).
[22]M. Hasegawa and M. Watabe, J. Phys. Soc. Jpn. **27**, 1393 (1969).
[23]K. Utsumi and S. Ichimaru, Phys. Rev. B **23**, 3291 (1981).
[24]P. K. Aravind, A. Holas, and K. S. Singwi, Phys. Rev. B **25**, 561 (1982).
[25]A. vomFelde, J. Sprösser-Prou, and J. Fink, Phys. Rev. B **40**, 10 181 (1989).
[26]K. J. Krane, J. Phys. F **8**, 2133 (1978).
[27]P. C. Gibbons, S. E. Schnatterly, J. J. Ritsko, and J. R. Fields, Phys. Rev. B **13**, 2451 (1976).
[28]K. S. Singwi, Phys. Scr. **32**, 397 (1985).
[29]W. C. Kerr, Phys. Rev. **174**, 316 (1969).
[30]F. Green, D. Neilson, and J. Szymanski, Phys. Rev. B **31**, 2779 (1985); **31**, 2796 (1985); **31**, 5837 (1985); **35**, 124 (1987); **35**, 133 (1987).
[31]M. Rösler and J. Devooght *et al.*, *Particle Induced Electron Emission I,* Springer Tracts in Modern Physics Vol. 122 (Springer-Verlag, Berlin, 1991).
[32]J. E. Borovsky and D. M. Suszcynsky, Phys. Rev. A **43**, 1433 (1991).
[33]K. Elk and W. Gasser, *Die Methode der Greenschen Funktionen in der Festkörperphysik* (Akademie-Verlag, Berlin, 1979).