First-principles calculation of the dynamic structure factor for the electron gas in metallic systems

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(Received 21 November 1984)

We have extended our previous microscopic numerical calculations for the dynamic structure factor of the electron gas at large momentum transfers to include the overall relaxation of the central peak due to electron-hole correlations and self-energies. These contributions vary slowly with energy transfer as had been anticipated. We confirm our excellent agreement with the multiple peaks experimentally observed in Li. Satisfaction of the sum rules is built into the formalism, and we confirm this numerically.

I. INTRODUCTION

In a series of papers we have formulated and applied[1–4] a new microscopic theory of the interacting electron gas at metallic densities. The theory can be validly used over the complete range of momentum transfer $\mathbf{q}$ and energy transfer ω. It treats static and dynamic correlations in a unified way and strictly conserves particle number, momentum, and energy.

In Refs. 3 and 4 we examined contributions to the structure factor $S(\mathbf{q},\omega)$ for large momentum transfers $q > k_F$ (k_F is the Fermi momentum) and for metallic densities greater than $r_s = 4$. We found that there are two quite distinct types of corrections to the random-phase approximation[5] (RPA) for the structure factor. Furthermore, these two types of terms can be treated as approximately independent of each other.

The first type of term was labeled class A in Ref. 4. These terms involve strong Coulomb scattering between pairs of excited electrons or holes. It was found that these terms alone lead to fine scale peaks in $S(\mathbf{q},\omega)$ as a function of ω for fixed $\mathbf{q}$. The other type of term, denoted as classes B, C, and D in Ref. 4, actually include contributions larger than the class-A terms, but these contributions all vary smoothly with ω and contain no fine-scale ω structure. They account for most of the overall relaxation of the main peak in $S(\mathbf{q},\omega)$.

It is interesting to note that the class-B, -C, and -D terms can be well approximated by a standard local-field construction in which one averages over hole momenta.[6] However, if one attempts to carry out an analogous approximation for the class-A terms, one finds that all the fine-scale peaks in ω become smeared out as a consequence of averaging, and one is left with an $S(\mathbf{q},\omega)$ consisting of a single smooth peak.

There are several advantages in treating the class-A terms quite separately from the other terms. First, one can demonstrate clearly that the observed fine-peak structure can be directly associated with this class of terms. Second, as noted, all except the class-A terms can be well approximated by a local-field construction, meaning that the approximations that can be validly employed are markedly different for the two types of terms. Third, our published results for the class-A contributions can be readily incorporated into any independent calculation of the other correction terms to the RPA. Of these corrections it is the Hartree-Fock terms[7] which are the most important, at large values of the momentum transfer $\mathbf{q}$.

The main disadvantage of including only the class-A terms is that a minor ambiguity is encountered when one wants to compare the position of the calculated peaks in $S(\mathbf{q},\omega)$ with the experimentally observed multipeak structure; in retaining only the class-A terms one neglects almost all the overall relaxation effects, so that the centroid of the resulting main peak will, by construction, coincide with that of the RPA peak instead of being centered on the observed peak. For the momentum-transfer range $1.5 < q/k_F < 3$, it is well known that the RPA peak is located about $2\epsilon_F$ above the fully relaxed peak (ϵ_F is the Fermi energy). Thus in comparing the calculated peaks with the observed structure we must rigidly shift all our curves in Refs. 2–4 towards lower ω by approximately $2\epsilon_F$. Since this almost rigid shift is nearly the same for all $\mathbf{q}$ in our range, only a single parameter is introduced. Furthermore the value of this energy-shift parameter is independently known to be approximately $2\epsilon_F$. Introducing this single shift in energy leads to good agreement between theory and experiment.[4]

In the present paper we incorporate the dominant class-B, -C, and -D contributions into our calculation, thus removing this remaining uncertainty.

II. THEORY

A. Hartree-Fock local field

The largest single contributions to the overall relaxation of the RPA peak in $S(\mathbf{q},\omega)$ at large $\mathbf{q}$ come from the Hartree-Fock series of terms for a particle-hole pair[7] (Fig. 1).

At large $\mathbf{q}$ all other terms contributing to the overall relaxation tend to be suppressed in comparison. This is because the dynamic dielectric function behaves like $1+0(k_F/q)$ when the characteristic energy transfers are of order $qk_F \gg \omega_p$ (ω_p is the plasma frequency). Therefore, only the leading Coulomb terms survive in both the

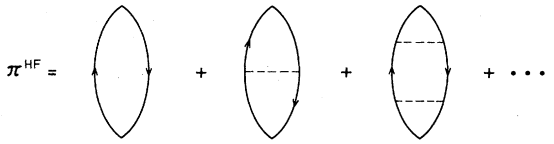

FIG. 1. Hartree-Fock contribution $\Pi^{HF}(\mathbf{q},\omega)$ to the polarization function $\Pi(\mathbf{q},\omega)$. The solid lines represent self-consistent single propagators G and the dotted horizontal lines are bare Coulomb interactions V.

particle-hole interactions and the single-particle self-energies. These leading terms are the Hartree-Fock contributions.

The remaining unscreened terms are small, simply because they involve Coulomb interactions through which momentum $\mathbf{q}$ must be transferred. Each such interaction contributes a factor of $1/q^2$. (In Ref. 4 we concentrated on the class-A terms even though they are small because they alone contain the fine scale structure effects.)

Dharma-wardana and Taylor[7] have examined the terms in Fig. 1 and calculated the fully iterated Hartree-Fock ladder series of unscreened Coulomb interactions between a particle and its hole. In their approach one writes down the integral equation for the full Hartree-Fock proper polarization function $\Pi^{HF}(\mathbf{q},\omega)$ and then approximately solves it using a straightforward iterative procedure.

The proper Hartree-Fock polarization function is given by[7]

$$\Pi^{HF}(\mathbf{q},\omega)=-\sum_{\mathbf{k}} P(\mathbf{k},\mathbf{q},\omega)\ , \tag{1}$$

where the vertex function $P(\mathbf{k},\mathbf{q},\omega)$ is the solution of the integral equation

$$P(\mathbf{k},\mathbf{q},\omega)=\frac{n(\mathbf{k},\mathbf{q})}{D^{(0)}(\mathbf{k},\mathbf{q},\omega)}-\sum_{\mathbf{k}_1} V(\mathbf{k}-\mathbf{k}_1)\left[\frac{n(\mathbf{k},\mathbf{q})}{D^{(0)}(\mathbf{k},\mathbf{g},\omega)}P(\mathbf{k}_1,\mathbf{q},\omega)-\frac{n(\mathbf{k}_1,\mathbf{q})}{D^{(0)}(\mathbf{k}_1,\mathbf{q},\omega)}P(\mathbf{k},\mathbf{q},\omega)\frac{D^{(0)}(\mathbf{k}_1,\mathbf{q},\omega)}{D^{(0)}(\mathbf{k},\mathbf{q},\omega)}\right]\ . \tag{2}$$

Here $V(\mathbf{k})=4\pi e^2/k^2$ is the bare Coulomb interaction, $n(\mathbf{k},\mathbf{q})=n_{\mathbf{k}}-n_{\mathbf{k}+\mathbf{q}}$ where $n_{\mathbf{k}}$ is the particle occupation number in the state $\mathbf{k}$. The denominator function is

$$D^{(0)}(\mathbf{k},\mathbf{q},\omega)=\omega+\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}+\mathbf{q}}\ , \tag{3}$$

where $\epsilon_{\mathbf{k}}=\hbar^2k^2/2m$ is the single-particle kinetic energy.

The fully iterated solution of Eq. (2) may be written

$$P(\mathbf{k},\mathbf{q},\omega)=n(\mathbf{k},\mathbf{q})/D(\mathbf{k},\mathbf{q},\omega)\ , \tag{4}$$

where the denominator $D(\mathbf{k},\mathbf{q},\omega)$ is the solution of another integral equation

$$D(\mathbf{k},\mathbf{q},\omega)=D^{(0)}(\mathbf{k},\mathbf{q},\omega)+\sum_{\mathbf{k}_1} V(\mathbf{k}-\mathbf{k}_1)n(\mathbf{k}_1,\mathbf{q})\left[\frac{D(\mathbf{k},\mathbf{q},\omega)}{D(\mathbf{k}_1,\mathbf{q},\omega)}-1\right]\ . \tag{5}$$

One obtains a self-consistent approximation for the solution of this equation by replacing the interaction $V(\mathbf{k}-\mathbf{k}_1)$ by $\langle V\rangle$, its average over the Fermi surface.[7,8]

With this approximation Eq. (5) can be solved for $D(\mathbf{k},\mathbf{q},\omega)$, from which the self-consistent Hartree-Fock polarization $\Pi^{HF}(\mathbf{q},\omega)$ can be determined using Eqs. (1) and (4).

Dharma-wardana and Taylor demonstrated that to a good approximation their fully iterated $\Pi^{HF}(\mathbf{q},\omega)$ could be represented by a simple expression

$$\begin{aligned}\mathrm{Re}\Pi^{HF}(\mathbf{q},\omega)&\simeq\frac{\mathrm{Re}\Pi^{0}(\mathbf{q},\omega)}{1+V(k_F)\,\mathrm{Re}\Pi^{0}(\mathbf{q},\omega)/4}+\frac{V(k_F)}{4}\left[\frac{\mathrm{Im}\Pi^{0}(\mathbf{q},\omega)}{1+V(k_F)\,\mathrm{Re}\Pi^{0}(\mathbf{q},\omega)/4}\right]^2,\\ \mathrm{Im}\Pi^{HF}(\mathbf{q},\omega)&\simeq\frac{\mathrm{Im}\Pi^{0}(\mathbf{q},\omega)}{[1+V(k_F)\,\mathrm{Re}\Pi^{0}(\mathbf{q},\omega)/4]^2}\ ,\end{aligned} \tag{6}$$

where $\Pi^0(\mathbf{q},\omega)$ is the familiar Lindhard polarization function[9] for a noninteracting particle-hole excitation.

Using the standard local-field construction the Hartree-Fock vertex function can be written

$$\Lambda^{HF}(\mathbf{k}_1,k_1^0;\mathbf{q},\omega)=\frac{\Lambda^0(\mathbf{k}_1,k_1^0;\mathbf{q},\omega)}{1+V(\mathbf{q})G(\mathbf{q},\omega)\Pi^0(\mathbf{q},\omega)}\ , \tag{7}$$

where $G(\mathbf{q},\omega)$ is a dynamic and complex local field given by

$$G(\mathbf{q},\omega)=V^{-1}(\mathbf{q})[\Pi^0(\mathbf{q},\omega)^{-1}-\Pi^{HF}(\mathbf{q},\omega)^{-1}]\ , \tag{8}$$

and $\Lambda^0(\mathbf{k}_1,k_1^0;\mathbf{q},\omega)$ is the noninteracting particle-hole vertex corresponding to $\Pi^0(\mathbf{q},\omega)$.

B. Dynamic structure factor

We now combine the results we have obtained for the static and approximately local contributions to $S(\mathbf{q},\omega)$ with the contributions of the class-A terms previously determined in Refs. 3 and 4. Using the notation of Ref. 4 we identify the approximately local part of the vertex function with its Hartree-Fock counterpart

$$\Lambda^{\mathrm{loc}}[G](\mathbf{k}_1,k_1^0;\mathbf{q},\omega)=\Lambda^{HF}(\mathbf{k}_1,k_1^0;\mathbf{q},\omega)\ . \tag{9}$$

Next we recall the effective interaction for pairs of particles and pairs of holes

$$\Xi^{\rm nl}_{\sigma_1\sigma_2}[G](\mathbf{k}_1,k_1^0,\mathbf{k}_2,k_2^0,\mathbf{q},\omega)=\langle \mathbf{q}+\mathbf{k}_1,\sigma_1,\mathbf{k}_2,\sigma_2 \mid \Xi^{e\text{-}e}(\omega+\epsilon_{\mathbf{k}_1}+\epsilon_{\mathbf{k}_2}) \mid \mathbf{k}_1,\sigma_1,\mathbf{q}+\mathbf{k}_2,\sigma_2\rangle$$
$$+\langle \mathbf{q}+\mathbf{k}_1,\sigma_1,\mathbf{k}_2,\sigma_2 \mid \Xi^{h\text{-}h}(\omega+\epsilon_{\mathbf{k}_1}+\epsilon_{\mathbf{k}_2}) \mid \mathbf{k}_1,\sigma_1;\mathbf{q}+\mathbf{k}_2,\sigma_2\rangle\ , \tag{10}$$

where σ is the spin index. The electron-electron effective interaction consists of a pair of dynamically screened interactions together with the infinite ladder sum of unscreened Coulomb interactions starting at third order.

When the momentum q is large compared to k_F, we find that the dynamic screening terms in $\Xi^{e\text{-}e}$ are suppressed to order k_F/q in comparison with the corresponding Coulomb terms.[4] Therefore, at large q, we approximate $\Xi^{e\text{-}e}$ by (see Fig. 2)

$$\Xi^{e\text{-}e}(\omega)=T^{e\text{-}e}(\omega)-V\ , \tag{11}$$

where $T^{e\text{-}e}(\omega)$ is the dynamic T matrix, defined as the solution of the Bethe-Goldstone ladder equation

$$T^{e\text{-}e}(\omega)=V+V\frac{Q^{>}}{\omega+\sum_h \epsilon_h-\sum_e \epsilon_e+i\eta}T^{e\text{-}e}(\omega)\ . \tag{12}$$

The subscript h on the kinetic energies ϵ, denotes hole states while e denotes electron states. $Q^{>}$ is the Pauli projection operator[6] ensuring that intermediate excitations are above the Fermi level. The hole-hole effective interaction $\Xi^{h\text{-}h}$ is defined analogously to Eqs. (11) and (12), with $Q^{<}\equiv(1-Q^{>})$ replacing $Q^{>}$.

The proper polarization function $\chi^{\rm sc}(\mathbf{q},\omega)$ can be expanded in powers of $\Xi^{\rm nl}$:

$$\chi^{\rm sc}(\mathbf{q},\omega)=2\sum_{\mathbf{k}_1}\int\frac{dk_1^0}{2\pi i}\Lambda^{\rm loc}[G](\mathbf{k}_1,k_1^0;\mathbf{q},\omega)+4\sum_{\mathbf{k}_1}\sum_{\mathbf{k}_2}\int\frac{dk_1^0}{2\pi i}\int\frac{dk_2^0}{2\pi i}\Lambda^{\rm loc}[G](\mathbf{k}_1,k_1^0;\mathbf{q},\omega)$$
$$\times\tfrac{1}{4}\sum_{\sigma_1,\sigma_2}\Xi^{\rm nl}_{\sigma_1,\sigma_2}[G](\mathbf{k}_1,\mathbf{k}_2k_2^0;\mathbf{q},\omega)\Lambda^{\rm loc}[G](\mathbf{k}_2,k_2^0;\mathbf{q},\omega)+\cdots\ , \tag{13}$$

where the truncated terms are of higher order in $\Xi^{\rm nl}[G]$.

The structure factor is then

$$S(\mathbf{q},\omega)=-\frac{1}{\pi}\,\mathrm{Im}\left[\frac{\chi^{\rm sc}(\mathbf{q},\omega)}{1-V(\mathbf{q})\chi^{\rm sc}(\mathbf{q},\omega)}\right]\ . \tag{14}$$

III. RESULTS AND DISCUSSION

In Fig. 3 we show our $S(\mathbf{q},\omega)$ calculated for $r_s=3.2$, and the structure factor data of Priftis, Boviatsis, and Vradis[10] for lithium. Both functions are plotted for fixed q as functions of ω. Since we have included relaxation effects in the present calculation, the curves may be compared directly.

We note that within experimental uncertainty there is a marked correspondence between the pairs of curves over the range $1.8<q/k_F<3$. As in Refs. 3 and 4, we find no evidence in our calculations of any peak or shoulder located around $\omega/\epsilon_F=2.5$ in the experimental observations at

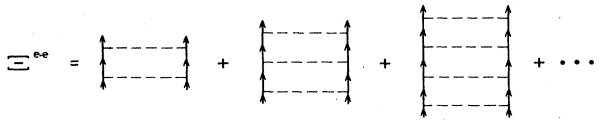

FIG. 2. Contributions to the electron-electron effective interaction $\Xi^{e\text{-}e}$. The horizontal dashed lines represent the bare Coulomb interaction V, the solid lines are the self-consistent dressed single-particle propagators G. The intermediate propagators are restricted to be above the Fermi surface.

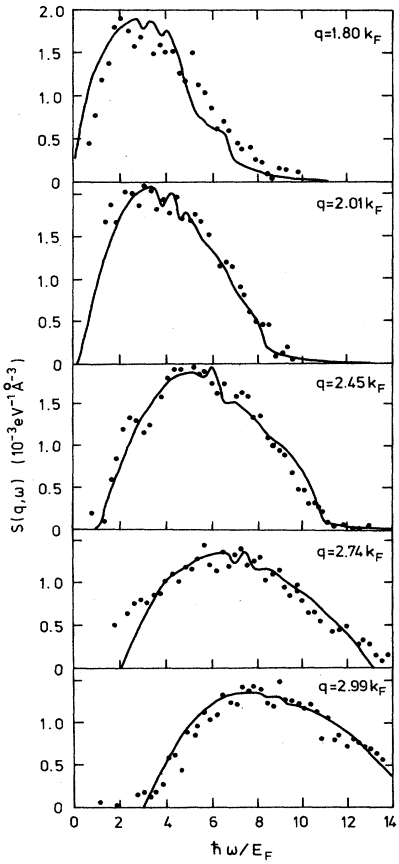

FIG. 3. Dynamic structure factor $S(\mathbf{q},\omega)$ (solid line) calculated for density $r_s=3.2$. The points are the experimental data from Ref. 10 for lithium.

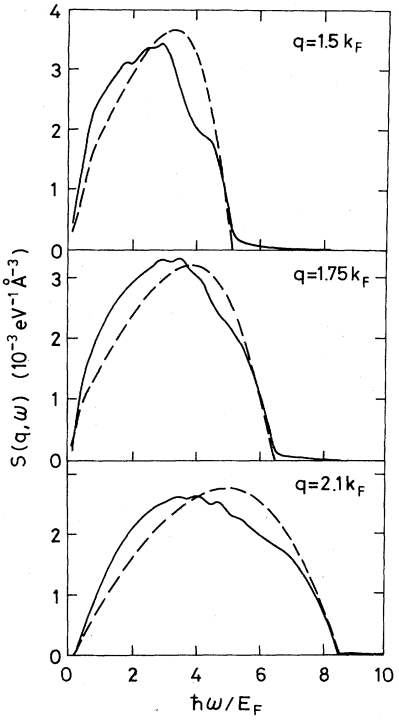

FIG. 4. Dynamic structure factor (solid line) for $r_s=2$ compared with the RPA result (dashed line).

$q/k_F=2.45$ and 2.74. We have argued that kinematic constraints exclude the possibility of a simple two-plasmon excitation in this region. We have previously speculated[4] that a possible mechanism for this peak would be in the excitation of a single plasmon accompanied by an interband electron transition assisted by an umklapp. This explanation is consistent with the peak's position and furthermore would only operate over the narrow momentum transfer range $2.3 < q/k_F < 3$, as observed.

In Fig. 4 we present our calculated results for $S(\mathbf{q},\omega)$ at $r_s=2$. As previously noted[1,11] it is difficult to use the published data of Platzman and Eisenberger[12] for Be in any direct comparison with our calculated curves for the following reason. If one assumes that only electron-gas effects contribute to their results in Ref. 12, it is straightforward to show that the data contain an internal inconsistency. The experimental $S(\mathbf{q},\omega)$ curves are not normalized, but one can normalize them so that the f-sum rule is exactly satisfied. One can then immediately obtain the static structure factor $S(q)$ which markedly differs from the $S(q)$ obtainable[11] from Monte Carlo data. For example, at $q/k_F=1.76$ the value of $S(q)$ from the Platzman-Eisenberger $S(\mathbf{q},\omega)$ is compared with the Monte Carlo value of 0.95 (our results give 0.96). A possible explanation of this inconsistency is that lattice effects are significantly contributing to their experimental results.

Although we have employed a formalism[13] for which the sum rules are automatically satisfied, it is of interest to check the validity of our numerical approximations by observing to what extent the various sum rules are violated. We find the f-sum rule at $r_s=2$ is satisfied in our numerical results to better than 7%. This is consistent with our numerical accuracy which is of the order of 5%.

The relative peak positions in Figs. 3 and 4 are almost unaltered compared with our previous calculations,[1-4] although in some instances there are some small quantitative changes. This is a reflection of the influence of relaxation and also of an improvement in our numerical accuracy.

In conclusion, we note that incorporating the overall relaxation contributions into our calculation leads to structure factors which conform to the sum rules and which are in good agreement with the available experimental data. Our curves show a great deal of fine structure as a function of ω which is consistent with the observed data. It would be useful to have measurements performed with better resolution to completely test our detailed predictions. The effects we have analyzed become more pronounced as the density is lowered. It would be of interest to have data at densities below $r_s=3.2$ for momentum transfers $q/k_F>2$.

ACKNOWLEDGMENTS

We wish to thank Roger Taylor for useful discussions, and for drawing our attention to the Fermi surface average approximation. This work was supported by a grant from the Australian Research Grants Scheme.

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