Structure factor of a correlated nonuniform fermion gas at small wavelength

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The structure factor of a nonuniform interacting electron gas is examined at large wave vectors to *delineate* the difference between lowest- and higher-order perturbation contributions. Implications for the exchange-correlation energy are also discussed.

Hubbard[1] and Nozières and Pines[2] were some of the first to recognize the utility of studying the detail dependence of the structure factor, for a uniform correlated electron gas, in terms of the individual wave vectors q. Following these ideas there were several recent extensions[3-8] of such calculations to nonuniform systems with some surprising results[8] (Ref. 8 will be referred to as I). I showed that any nonuniformity, superimposed on a uniform Fermion system, *fundamentally* changes the nature of the structure factor $S_\lambda(\vec{r},\vec{r}')$ in the *thermodynamic* limit. The $S_\lambda(\vec{r},\vec{r}')$ or equivalently the exchange-Coulomb hole[7,8] is given by

$$S_\lambda(\vec{r},\vec{r}')=\langle\phi_\lambda|[\hat{\rho}(\vec{r})-\rho(\vec{r})][\hat{\rho}(\vec{r}')-\rho(\vec{r}')]|\phi_\lambda\rangle \ , \quad (1)$$

where ϕ_λ is the full ground state for the nonuniform interacting Fermion system with interparticle interaction $\lambda v(\vec{q})$ and $\hat{\rho}(\vec{r})$ is the density operator. [Concerning $v(q)$ we take the form $\lambda v(q)=\lambda 4\pi e^2/(q^2+\alpha^2)$.]

We summarize the conclusions of I:

(i) For Hartree-Fock (HF) *alone* a metal surface introduces long-range behavior in the exchange-hole such that

$$\lim_{\vec{q}\to 0} S_{\lambda=1}(\vec{q},\vec{q})\equiv \lim_{\vec{q}\to\vec{0}}\int d^3r\int d^3r' e^{i\vec{q}\cdot(\vec{r}-\vec{r}')}S_{\lambda=1}(\vec{r},\vec{r}')\neq\int d^3r\int d^3r' S_{\lambda=1}(\vec{r},\vec{r}')=0 \ . \quad (2a)$$

The last equality is simply a statement of the conservation of particles [Eq. (1)].

The connection between $S_\lambda(\vec{q},\vec{q})$ and the exchange-correlation energy is[4,5]

$$E_{xc}=\int\frac{d^3q}{(2\pi)^3}E_{xc}(\vec{q})$$
$$=N\tfrac{1}{2}\int\frac{d^3q}{(2\pi)^3}v(\vec{q})\int_0^1 d\lambda\,[S_\lambda(\vec{q},\vec{q})-1] \quad (2b)$$

which implies similar subtlety for $E_{xc}(\vec{q})$ at small $\vec{q}$ for long-range $v(q)$ (i.e., $\alpha\to 0$; see below).

(ii) The long-range part of the exchange-Coulomb hole (or equivalently the small-q limit) of a weakly nonuniform fully correlated electron gas is *extremely sensitive* to the external perturbation. Consequently, the small q region of $E_{xc}(q)$ is modified by the detail density profile and interpolations, in that range, are clearly inappropriate.

(iii) The behavior of Eq. (2a) at a metal surface is modified only for a Coulomb interparticle interaction $v(\vec{q})$ ($\alpha\to 0$, see also Refs. 9–11). For such a long-range interaction the term responsible for the behavior of Eq. (2a) is screened and the small-q behavior is now governed by the detail of the surface electronic profile, in agreement with (ii) (see also Ref. 10). For example, the E_{xc} contribution $k_F 8\pi q(\omega_S-\omega_B/2)$ (where ω_B and ω_S are the bulk and surface plasmons,[4] respectively) to the surface energy at small q appropriate to the *semiclassical* approximation[12-14] is now modified by the detail surface density profile.

The above three results pertain to the long-wavelength limit (small q) of $S_{\lambda=1}(\vec{q},\vec{q})$. For large q I only briefly examined the contention that the corresponding $E_{xc}(q)$ is given exactly in the local-density approximation (LDA).[4] In this Brief Report we examine the expected differences from higher-order perturbation.

It will turn out that it is sufficient to consider the structure factor $S_\lambda(\vec{q},\vec{q})$ to second order in an external potential $V_b(\vec{r})$ of a single Fourier component $\vec{k}$ (i.e., $V_b(\vec{r})=V_b(\vec{k})e^{i\vec{k}\cdot\vec{r}}$). To first order in the interparticle interaction, the diagrams for the structure factor are presented in Fig. 1. We exclude HF contributions to $S_\lambda(\vec{q},\vec{q})$ (some examples are shown in Fig. 2) since these graphs are known to give *identically* zero contribution above $q=2k_F$.[5,6] In the limit of $q\to\infty$ and after some algebra required to evaluate the Feynman diagrams of Figs. 1 and 2 the leading contributions to $S_\lambda(\vec{q},\vec{q})$ can be extracted. The results are as follows:

(a) Graphs (1) and (2) of Fig. 1 and (8) of Fig. 2 make the following leading contribution in $1/q$,

$$\lim_{q\to\infty} S_\lambda(\vec{q},\vec{q})=\frac{A\lambda e^2}{q^4}\Pi_0^2(\vec{k})V_b^2(\vec{k}) \ , \quad (3a)$$

where A is a constant *independent* of $\vec{q}$, $\vec{k}$, and e^2. $\Pi_0(\vec{k})$ is the usual Lindhard screening function

$$\Pi_0(\vec{k})=\int\frac{d^3p}{(2\pi)^3}\frac{\Theta^<(\epsilon_{\vec{p}})-\Theta^<(\epsilon_{\vec{p}+\vec{k}})}{\epsilon_{\vec{p}+\vec{k}}-\epsilon_{\vec{p}}} \ , \quad (3b)$$

where $\epsilon_p=p^2/2m-\epsilon_F$ and $\Theta^<(x)=1$ for $x<0$ and $\Theta^<(x)=0$ for $x>0$.

(b) Graphs (5)–(8) of Fig. 1 give

$$\lim_{q\to\infty} S_\lambda(\vec{q},\vec{q})=\frac{B\lambda e^2}{q^4}\Pi_1(\vec{k})V_b^2(\vec{k}) \ , \quad (4a)$$

where again B is a constant independent of $\vec{q}$, $\vec{k}$, and e^2 and

$$\Pi_1(\vec{k})=\int\frac{d^3p}{(2\pi)^3}\frac{[\Theta^>(\epsilon_{\vec{p}})\Theta^<(\epsilon_{\vec{p}+\vec{k}})-\Theta^<(\epsilon_{\vec{p}})\Theta^>(\epsilon_{\vec{p}+\vec{k}})]}{(\epsilon_{\vec{p}+\vec{k}}-\epsilon_{\vec{p}})^2}=0 \ , \quad (4b)$$

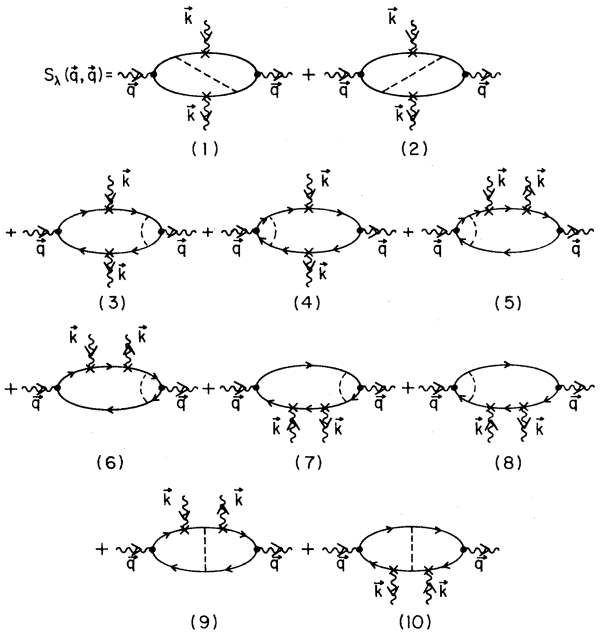

FIG. 1. *All* lowest-order *non*-Hartree-Fock contributions to the structure factor $S_\lambda(\vec{q},\vec{q})$. The wiggly line ending with the cross is the bare external perturbation $V_b(\vec{k})$. The solid arrowed lines are the electron propagators and the dashed lines are the interparticle interaction λv.

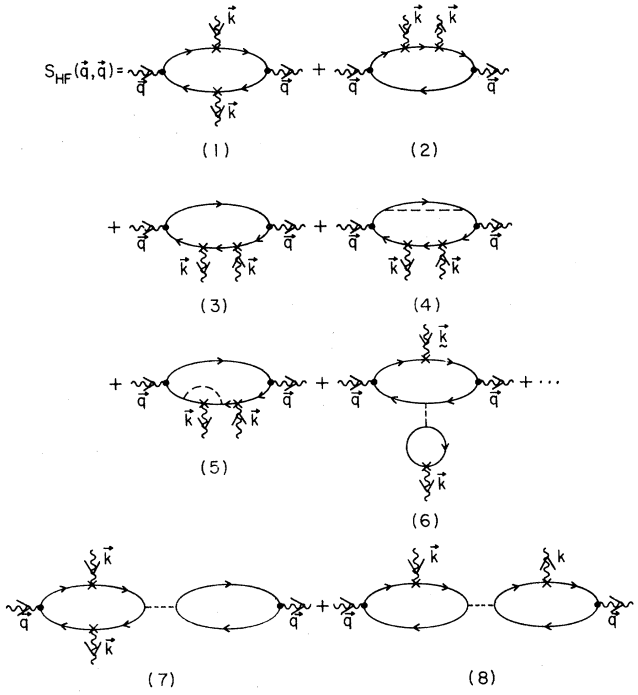

FIG. 2. Examples of Hartree-Fock [(1)–(6)] or RPA [(7),(8)] contributions to the structure factor.

where $\Theta^{>}(x)=1-\Theta^{<}(x)$.

(c) Graphs 3, 4, 9, and 10 of Fig. 1 and the HF or random-phase-approximation (RPA) contributions [see Fig. 2, (1)–(7)] are either identically zero or make higher-order contributions in $1/q$. The structure factor for a uniform interacting electron gas goes like $1/q^4$ at large q (Refs. 1 and 2) and our results show that nonuniform corrections carry the same leading terms in $1/q$.

We next turn to the implication of the above results for the E_{xc} in the LDA, at *large q*. To make the connection with E_{xc} in the LDA and Eq. (2b) we recall that the contributions of Figs. 1 and 2 in conjunction with Eq. (2b) contain the term $\int d^3r\, V_b(\vec{r})\rho(\vec{r}) = V_b(\vec{k})\rho(\vec{k})$. This term is *not* part of the E_{xc} contribution in the density functional formalism (FD) and must be subtracted out. Upon integration over the coupling constant and making the above subtraction we get for the lowest-order *nonuniform* contribution to E_{xc} in the FD

$$\lim_{q\to\infty} E_{xc}(\vec{q}) = -\frac{1}{4}\frac{v(q)e^2}{q^4} V_b^2(\vec{k}) A\, \Pi_0^2(\vec{k}) \quad . \tag{5}$$

It is preferable to replace $V_b(\vec{k})$ by the induced density $\rho(\vec{k})$. To *lowest order* in e^2,

$$\rho(\vec{k}) = -\Pi_0(\vec{k})\, V_b(\vec{k}) \tag{6a}$$

and

$$\lim_{q\to\infty} E_{xc}(q) = -\frac{1}{4}\frac{v(q)e^2}{q^4}\rho^2(\vec{k})A \quad . \tag{6b}$$

Since A is *independent* of $\vec{k}$ Eq. (6b) is accounted for to lowest order *entirely* in the LDA in agreement with the previous conclusions.[4] This result *hinges* on the structure of Eqs. (3a) and (6a) which both contain $\Pi_0(\vec{k})$ in lowest-order perturbation.

We now see the difficulty of extending the above conclusions to higher order where such a simple relation is no longer valid, and where terms also make a $1/q^4$ contribution, as can be verified by direct evaluation of the higher-order Feynman graphs. We conclude that the result of the lowest-order perturbation theory cannot be trivially extended to higher order and the contention that the $\lim_{q\to\infty} E_{xc}^{LDA}(q)$ is exact remains in question.

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