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# DYNAMIC CORRELATIONS IN THE ELECTRON GAS.°

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Abstract.

For the calculation of many properties of simple metals, the "jellium" model is widely used as a first approximation. The frequency- and wave vector-dependent dielectric function of this model not only allows to study its dielectric response, but also to calculate several other properties, e.g. the ground state energy, the density-fluctuation excitation spectrum, the pair correlation function, ...

In the last decade, experimental and theoretical evidence revealed the need for a frequency-dependent description of exchange and correlation effects. In the present paper, we review a method for introducing these effects starting from the equation of motion for the Wigner distribution function in the presence of a weak external perturbation. The application of the Hartree-Fock decoupling in this equation of motion leads to an integro-differential equation, which is studied with a variational approach, leading to a frequency dependent "local-field correction" $G(q,\omega)$. The dynamical exchange effects substantially influence the dielectric function. For instance, compared to RPA, the plasmon frequencies in the particle-hole continuum are appreciably lowered. This tendency is confirmed experimentally. Furthermore, in contrast to most other approximations, this method satisfies a large class of consistency requirements and sum rules, for which a dynamical treatment of exchange and correlation is required.

Several other approximations to include dynamical exchange effects in the dielectric function are discussed in the framework of the Wigner distribution function, which shows their interrelation in a transparent way. Most of these approaches are shown to be approximations of the variational result.

## I. Introduction.

The calculation of many properties of simple metals is based on the "jellium model". In this model, one merely studies the interaction among the electrons themselves, whereas the lattice of positive metal ions is replaced by a rigid uniform positive background. The large amount of research on this hypothetical system is not only due to its conceptual simplicity. The main reason for the persisting interest is the fact that in many real metals the conduction electrons can be approximately treated as having a homogeneous distribution in space. Furthermore, many theories for inhomogeneous systems, use the jellium model as a starting point for further investigations.

A basic quantity for the study of the jellium model is the frequency- and wave vector-dependent longitudinal dielectric function $\varepsilon(q,\omega)$, which not only allows the study of the dielectric response, but also the calculation of other properties, such as e.g. the ground state energy, the dynamic structure factor $S(q,\omega)$, the pair correlation function $g(r)$, ...

Since the pioneering work [1] of Lindhard, Bohm, Pines, Nozières, ..., the dielectric function in the so-called Random Phase Approximation (RPA) has been the standard reference basis for further improvements. In this approximation, one calculates the response of the electrons subjected to an external field, assuming that each electron moves in the Hartree field of all the other electrons. In the long-wavelength limit, the RPA succeeds in explaining basically the dispersion of the energy of the collective excitations, and simultaneously to combine the ideas of screening, collective excitations and single-particle excitations. However, despite its merits, the RPA is less satisfactory whenever short-range correlations are important. For instance, energy loss measurements of fast electrons and X-ray scattering experiments revealed appreciable deviations in the structure factor from the RPA predictions [2-8]. This failure of the RPA in the short-range description is also revealed by the fact that for metallic densities the pair correlation function becomes negative near the origin [9].

A large variety of approximations has been proposed to improve upon the RPA. In these approximations one calculates the "local field correction" $G(q,\omega)$ occurring in the dielectric function, which is usually written in the form:

$$\varepsilon(q,\omega) = 1 + \frac{Q_o(q,\omega)}{1-G(q,\omega)Q_o(q,\omega)} \tag{1}$$

In this expression, $Q_o(q,\omega)$ is the Lindhard function:

$$Q_o(q,\omega) = \frac{4\pi e^2}{q^2} \int d^3p \, \frac{N_{\vec{q}}(\vec{p})}{\omega^+ - \vec{p}.\vec{q}/m} \tag{2}$$

where $\omega^+ = \omega + i\varepsilon$ accounts for the adiabatic switching of external perturbations, and $N_{\vec{q}}(\vec{p})$ is a geometrical factor, related to the equilibrium momentum distribution function $f^o(p)$ of the electron gas:

$$N_{\vec{q}}(\vec{p}) = \frac{1}{\hbar}\left[ f^o(\vec{p}+\hbar\vec{q}/2) - f^o(\vec{p}-\hbar\vec{q}/2)\right] \tag{3}$$

The Lindhard function $Q_o(q,\omega)$ is analytically known [1], and it determines the RPA dielectric function:

$$\varepsilon_{RPA}(q,\omega) = 1 + Q_o(q,\omega) \tag{4}$$

The function $G(q,\omega)$ describes the exchange and correlation potential on each electron due to other electrons. Originally, not much effort was done to include the frequency dependence in $G(q,\omega)$, and most studies concentrated on static approximations $G(q)$ [10-22], which were then often used in the dynamic dielectric function. The interest in the explicit frequency dependence of $G(q,\omega)$ was rather exceptional [23], and restricted to some limiting cases.

In the last decade however, the dynamics of the exchange and correlation hole became an important topic, in view

of the more accurate measurements of the dynamic structure factor at large wave vector [2-8], and because sum rules and causality arguments revealed that a static treatment of $G(q,\omega)$ necessarily leads to theoretical inconsistencies [24-26].

A variety of different approaches has been used in order to derive expressions for $G(q,\omega)$ (although not many explicit calculations of the full frequency- and wave vector-dependence have been performed). In the present paper, the equation of motion for the Wigner distribution function will be used, because of its physical transparency, and because it easily reveals the close connection between various approximations. This approach essentially excludes a detailed discussion in terms of the Mori formalism [27]. The latter method is particularly useful for studying the dynamics of a system with known static properties, since it guarantees by construction that the relevant sum rules in the high- and low-frequency limits are satisfied. The lack of detailed knowledge of the static properties of the electron liquid however, complicates the interpretation of the physical significance of the application of the Mori formalism in this domain [28-30].

## II. Decoupling of the equation of motion for the Wigner distribution function.

The Wigner distribution function $f_\sigma(\vec{p},\vec{R},t)$ is the quantum analogue of the classical Boltzmann distribution function for particles of spin $\sigma$ with momentum $\vec{p}$ to be found in position $\vec{R}$ at time t [31]. It is defined as

$$f_\sigma(\vec{p},\vec{R},t) = \frac{1}{(2\pi\hbar)^3}\int d^3r\, e^{-i\vec{p}.\vec{r}/\hbar}\,\langle\Psi^+_\sigma(\vec{R}-\frac{\vec{r}}{2})\Psi_\sigma(\vec{R}+\frac{\vec{r}}{2})\rangle_t \qquad (5)$$

where $\langle\ldots\rangle_t$ denotes the expectation value of the operators at time t, and $\Psi$, $\Psi^+$ are the usual annihilation and creation operators for fermions. Similar to classical mechanics, the density and current density are obtained from:

$$n(\vec{R},t) = \sum_\sigma \int d^3p\ f_\sigma(\vec{p},\vec{R},t)\ . \tag{6a}$$

$$j(\vec{R},t) = \sum_\sigma \int d^3p\ \frac{\vec{p}}{m}\ f_\sigma(\vec{p},\vec{R},t)\ . \tag{6b}$$

In the presence of an external scalar field with Fourier components $e\varphi_{\vec{q}\omega}$, the equation of motion for $f_\sigma(\vec{p},\vec{R},t)$ can be derived (using the standard anticommutation relation for fermions) from the commutator of $\psi^+_\sigma(\vec{x}')\psi_\sigma(\vec{x})$ with the hamiltonian. As a result one obtains an expression which contains the two-particle Wigner distribution function:

$$f^{(2)}_{\sigma\sigma'}(\vec{p},\vec{p}';\vec{R},\vec{R}',t) = \frac{1}{(2\pi\hbar)^6}\int d^3r \int d^3r'\ e^{-i\vec{p}.\vec{r}/\hbar}\ e^{-i\vec{p}'.\vec{r}'/\hbar}$$

$$<\psi^+_\sigma(\vec{R}-\frac{\vec{r}}{2})\psi^+_{\sigma'}(\vec{R}'-\frac{\vec{r}'}{2})\psi_{\sigma'}(\vec{R}'+\frac{\vec{r}'}{2})\psi_\sigma(\vec{R}+\frac{\vec{r}}{2})>_t \tag{7}$$

One can proceed by deriving the equation of motion for the two-particle Wigner distribution function and so on. Continuing this way, one finds the well-known BBGKY hierarchy, in which distribution functions of higher order enter successively. A detailed study of the two-particle distribution function has been made in [25].

The main problem is to find good approximations to break this hierarchy, in order to be able to calculate the Wigner distribution function, and subsequently the induced electron density. In the present paper we adopt the Hartree-Fock decoupling, written schematically as:

$$<\psi^+_1\psi^+_2\psi_3\psi_4> \simeq <\psi^+_1\psi_4><\psi^+_2\psi_3> - <\psi^+_1\psi_3><\psi^+_2\psi_4> \tag{8}$$

One then obtains the equation of motion:

$$\tilde{f}_\sigma(\vec{p},\vec{q},\omega) = \frac{-\frac{1}{2}N_{\vec{q}}(\vec{p})U_{\vec{q}\omega} + \tilde{X}_\sigma(\vec{p},\vec{q},\omega)}{\omega^+ - \vec{p}.\vec{q}/m} \tag{9}$$

where $U_{\vec{q}\omega}$ is the total Hartree field:

$$U_{\vec{q}\omega} = e\varphi_{\vec{q}\omega} + \frac{4\pi e^2}{q^2} \sum_\sigma \int d^3p \, \tilde{f}_\sigma(\vec{p},\vec{q},\omega) \tag{10}$$

and $\tilde{X}_\sigma$ describes the effects of the exchange interaction:

$$\tilde{X}_\sigma(\vec{p},\vec{q},\omega) = \frac{1}{2} \int d^3p \, \frac{4\pi e^2\hbar^2}{|\vec{p}-\vec{p}'|^2} \left[ N_{\vec{q}}(\vec{p}) \tilde{f}_\sigma(\vec{p}',\vec{q},\omega) - N_{\vec{q}}(\vec{p}') \tilde{f}_\sigma(\vec{p},\vec{q},\omega) \right] \tag{11}$$

$\tilde{f}_\sigma(\vec{p},\vec{q},\omega)$ is the space-time Fourier transform of $f_\sigma(\vec{p},\vec{R},t) - f^o_\sigma(\vec{p})$, i.e. the non-equilibrium contribution in the Wigner distribution function. Solving the integral equation (9-11) allows to calculate the dielectric function with dynamical exchange effects, since the induced electron density

$$n_{\vec{q}\omega} = \sum_\sigma \int d^3p \, \tilde{f}_\sigma(\vec{p},\vec{q},\omega) \ . \tag{12}$$

is related to the external field potential $e\varphi_{\vec{q}\omega}$ through the defining equation for the dielectric function:

$$e\varphi_{\vec{q}\omega} + \frac{4\pi e^2}{q^2} n_{\vec{q}\omega} = \frac{e\varphi_{\vec{q}\omega}}{\varepsilon(q,\omega)} \tag{13}$$

III. Comparison of various approximations.

Obviously, the exchange term (11) forms the complicating factor in solving the equation of motion (9). Indeed, if one neglects $\tilde{X}_\sigma$, the solution of (9) is easily obtained, leading to the Lindhard distribution function:

$$f^L_\sigma(\vec{p},\vec{q},\omega) = -\frac{1}{2} e\varphi_{\vec{q}\omega} \frac{1}{1+Q_o(q,\omega)} \frac{N_{\vec{q}}(\vec{p})}{\omega^+ - \vec{p}.\vec{q}/m} \tag{14}$$

Applying (12) and (13), one readily derives that this corresponds to the RPA dielectric function (4).

In an attempt to account for exchange effects, we have applied a variational technique in [32], which consists in deriving a functional $F[f_\sigma(\vec{p},\vec{q},\omega)]$ with the property that the equation of motion (9) follows from the extremum condition:

$$\frac{\delta F[f]}{\delta f_\sigma(\vec{p},\vec{q},\omega)} = 0 \tag{15}$$

Taking as a trial function

$$f_\sigma^{trial}(\vec{p},\vec{q},\omega) = f_\sigma^L(\vec{p},\vec{q},\omega)\gamma_{\vec{q}\omega} \tag{16}$$

with $\gamma_{\vec{q}\omega}$ independent of momentum p, an algebraic equation for $\gamma_{\vec{q}\omega}$ follows from (15). This equation is readily solved. The result for $G(q,\omega)$, obtained in [32] is:

$$G_{var}(q,\omega) = \frac{4\pi e^2}{q^2}\frac{2\pi e^2\hbar^2}{Q_o^2(q,\omega)}\int d^3p'\int d^3p\,\frac{N_{\vec{q}}(\vec{p})N_{\vec{q}}(\vec{p}')}{\omega^+ - \vec{p}.\vec{q}/m}\,\frac{1}{|\vec{p}-\vec{p}'|^2}\,x$$

$$x\left[\frac{1}{\omega^+ - \vec{p}'.\vec{q}/m} - \frac{1}{\omega^+ - \vec{p}.\vec{q}/m}\right] \tag{17}$$

Although (17) is a rather complicated sixfold integral, it could be reduced analytically to a double numerically tractable integral. To the best of our knowledge, (17) has been the first expression for $G(q,\omega)$ which was evaluated with respect to both the wave vector and frequency dependence. Details on the numerical and analytical techniques, figures and tables, and a more extensive discussion of the physical implications, can be found in [32-35]. The results are:

1) The continuity equation, and all other sum rules checked up to now are exactly satisfied.
2) $G(q,\omega)$ is a universal function of $q/k_F$ and $\omega/E_F$ for all densities. This seems to be confirmed by experimental energy loss spectra of fast electrons in aluminum and sodium [36], as illustrated in fig. 1.
3) The plasmon energy is substantially lowered compared to

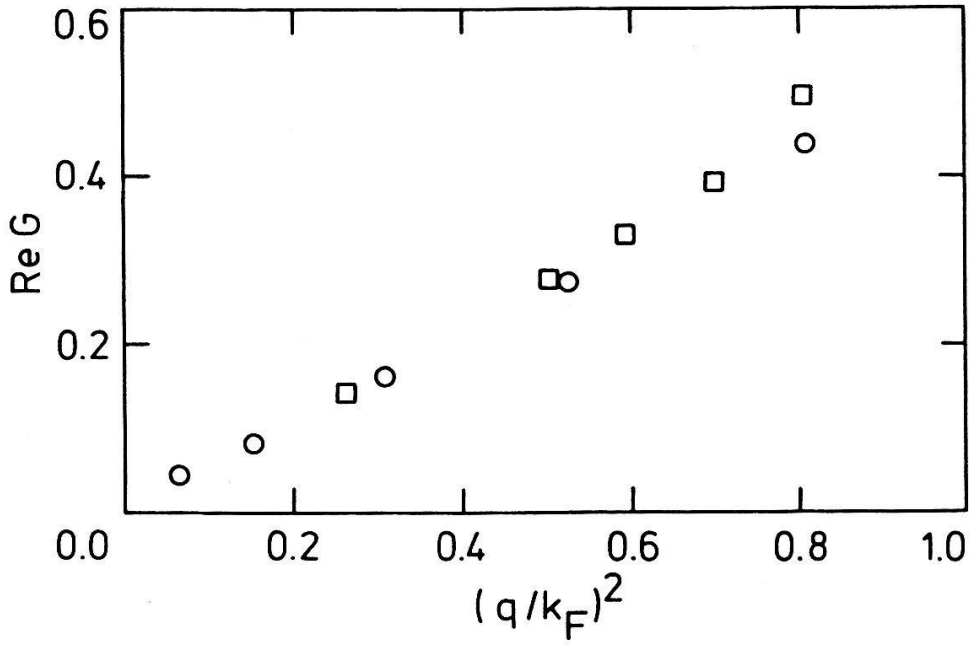


Fig.1. Fitted values of the real part of the local field correction for different wave vectors (expressed in units of the Fermi wave vector, at the frequencies of the experimental maxima [36] in the dynamical structure factor. The circles and the squares represent the fitted data for sodium ($r_s$ = 3.96) and aluminum ($r_s$ = 2.07) respectively.

RPA at large wave vector. In aluminum, this appreciably improves the agreement with experimental data [6].

We have also used this variational technique in quasi-one-dimensional systems, and dynamical exchange effects turned out to allow, at least qualitatively, to explain the anomalous plasmon dispersion of TTF-TCNQ [37]. For a quantitative analysis however, more detailed information on the band structure is required.

By a quite different method, the variational result (17) was later obtained by Tripathy and Mandal [38]. They applied the decoupling (8) in the equation of motion for the double-time-retarded commutator of the charge-density-fluctuation operators, and imposed conservation of frequency moments to all orders in the Hartree-Fock approximation for the static properties. Their method is an extension of an earlier approximation

by Toigo and Woodruff [39], who imposed only conservation of the first frequency moment. Unfortunately, the explicit evaluations of the dielectric function of [38] turned out to be seriously in error [40-43].

The integral equation (9) has also been studied by an iterative approach to first order in the exchange effects [23], starting from the Lindhard distribution function (14), and treating $\tilde{X}_\sigma$ as a perturbation. However, no explicit evaluation was performed. The result obtained to first order is

$$\varepsilon_{it}(q,\omega) = 1 + Q_o(q,\omega)\,[\,1 + G_{var}(q,\omega)Q_o(q,\omega)\,] \qquad (18)$$

Comparing (18) to (1), it is readily observed that the iterative dielectric function is the first order expansion of the variational dielectric function.

It is interesting to note that the static limit of (18) has been evaluated by Geldart and Taylor [18], by considering the diagrams for the proper polarizability to first order in the electron-electron interaction. This diagrammatic expansion has been evaluated dynamically by Holas, Aravind and Singwi [44], again yielding the iterative dielectric function (18). The main problem of (18) however is the fact that the imaginary part of the dielectric function becomes negative for certain values of q and $\omega$.

In this context, a different approach by Brener and Fry [45] should be mentioned, who iteratively treated a set of self-consistent equations for the self-energy, polarization and Green's function [16]. Their dielectric function can be obtained by the iteration scheme discussed above, but starting from the Lindhard distribution function (14) including the Hartree-Fock self-energy in the denominator $\omega^+ - \vec{p}.\vec{q}/m$. A disadvantage of this approach is that in each stage of the calculation the continuity equation is violated.

A rather controversial point in the treatment of dynamical exchange effects, is the fact that $G_{var}(q,\omega)$, as given in (17), is logarithmically divergent at the boundaries of the

particle-hole continuum $\omega = \hbar|qk_F \pm q^2/2|/m$. The physical significance of these singularities is not quite clear [33-35, 44]. Recently it has been shown that a generalization of the trial distribution function (16), including momentum corrections to order $p^2$, eliminates these logarithmic singularities [46], and leaves $G_{var}(q,\omega)$ essentially unaltered outside the singular regions. This is possibly an indication that the singularities might be an artefact of the approximations made, rather than having to do with the physics of the exchange hole.

## IV. Concluding remarks

The variational solution of the decoupled equation of motion (9-11) for the Wigner distribution might serve as a starting point for further studies of exchange and correlation in the dielectric function. Its connection to several other approaches has been examined, showing that many of them are particular cases or additional approximations to this variational approach. Furthermore, the improvement upon RPA from dynamical exchange effects, compared to experimental data, and the fact that all checked sum rules are satisfied, gives confidence in the variational and related approaches, even in studying the equation of motion for the two-particle distribution function [47].

Up to now, very few conclusions can be drawn on the dynamic correlations beyond the exchange decoupling. Even in the static limit, most recent studies make the approximation that $G(q,\omega)$ should not depend on frequency, although dynamical sum rules are implicitly imposed [48]. Also numerical methods up to now do not include the frequency dependence. For instance, the recent calculation by Lantto et al. [49] of the dielectric response using a Jastrow variational many-body wave function [50], explicitly assumes a static approximation $G(q)$ for $G(q,\omega)$. One of the consequences is that the large vector limit in [49] is not correctly related to the pair correlation function $g(r)$ in the origin. Also in the numerical treatments, one can hardly avoid the unpleasant complication of accepting the frequency dependence in $G(q,\omega)$.

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