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Time-dependent Hartree-Fock formalism for the dielectric function

N. E. Brener and J. L. Fry
University of Texas at Arlington, Arlington, Texas 76019

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In this paper we derive a formula for the time-dependent Hartree-Fock dielectric function by using Green's-function theory. We develop a set of self-consistent equations for the self energy, polarization, and Green's function which can be iterated to obtain successively more accurate approximations for the polarization. In this procedure the Hartree and Hartree-Fock approximations occur naturally as the first and second steps in the iterations. As the iterations are carried out, we obtain not only the expressions for the polarization, but also the one-electron Hartree-Fock equations, so that there is no question as to which polarization formula corresponds to which equation. In this way we obtain an expression for the dielectric function which clearly and directly corresponds to the Hartree-Fock equation and which is therefore called the time-dependent Hartree-Fock (TDHF) dielectric function. In this TDHF formula for the dielectric function, the first term is the time-dependent Hartree or random-phase approximation dielectric function. We show that in order to be consistent with the theory, one should use Hartree wave functions and energies to compute random-phase approximation dielectric functions and Hartree-Fock wave functions and energies to compute TDHF dielectric functions. Numerical results are presented for the free-electron gas.

I. INTRODUCTION

In recent years there has been a great deal of interest in calculating the dielectric response function which is a basic quantity used to determine many physical properties of solids. In most of these calculations the formula used for the dielectric function is the random-phase approximation (RPA)[1,2] which is derived from the time-dependent Hartree equation and thus contains no exchange terms. Since exchange effects play an important part in many physical processes, it is desirable to go beyond the RPA to include such effects in the dielectric function.

Attempts have been made by previous authors to go beyond the RPA, which is also known as the time-dependent Hartree approximation because of its direct relationship to the time-dependent Hartree equation. Hanke and Sham[3,4] have included an infinite sum of ladder diagrams in the polarization and have chosen to call this expression the time-dependent Hartree-Fock approximation. However, there is no direct correspondence between the infinite sum of ladder diagrams and the one-electron Hartree-Fock equation, and therefore it is questionable to use the term "time-dependent Hartree-Fock approximation" when referring to such an infinite sum. Also the sum of ladder diagrams is only a partial summation, including only one diagram in each order. There are many other diagrams besides the ladder diagrams that contribute to the polarization.

In this paper we take a different approach. Rather than attempt to make infinite partial summations of diagrams, we follow the method of Hedin and Lundqvist[5] and develop a set of self-consistent equations for the Green's function, self-energy, and polarization. We then iterate this set of equations to obtain successively more accurate approximations for the polarization. In this iterative procedure, the Hartree and Hartree-Fock approximations occur naturally as the first and second steps in the iterations. As these iterations are carried out, we obtain not only the expressions for the polarization but also the corresponding one-electron equations themselves, i.e., the Hartree and Hartree-Fock equations. Thus in this procedure there is no question as to which polarization expression corresponds to the Hartree equation and which one corresponds to the Hartree-Fock equation. In this way we obtain an expression for the dielectric function which clearly and directly corresponds to the Hartree-Fock equation and which is therefore called the time-dependent Hartree-Fock (TDHF) dielectric function. In the set of self-consistent equations used to obtain this TDHF formula for the dielectric function we have included not just the ladder diagrams but all of the diagrams that contribute to the polarization. The expression that we obtain in this manner has a completely different form from the result that one gets by summing ladder diagrams. Our Green's-function method yields an explicit formula for the time-dependent Hartree-Fock polarization that differs from the time-dependent Hartree polarization by the addition of a term containing exchange effects, just as the Hartree-Fock equation itself differs from the Hartree equation by the addition of an exchange term. On the other hand, the sum of ladder diagrams leads to an integral equation in which case the polarization cannot be written as the sum of the RPA term plus an additional term.

As indicated above, there are several expressions that one can use for the dielectric function, including the RPA and TDHF formulas. However, there appears to be a great deal of uncertainty in the literature as to which are the correct wave functions and energies to use in a particular dielectric function formula. While the RPA formula is in wide use, there is no uniformity of energies and wave functions used to compute it, but instead different authors have used a wide variety of different wave functions and energies, including those given by the Hartree,[6] Hartree-Fock,[7] correlated Hartree-Fock,[8] pseudopotential,[9] and Hartree-Fock-Slater[10,11] approximations. In this paper we show clearly that in order to be consistent with the theory, one should use Hartree wave functions and energies in the RPA formula and Hartree-Fock wave functions and energies in the TDHF formula.

II. SUMMARY OF GREEN'S-FUNCTION FORMALISM

Following Hedin and Lundqvist,[5] one can use Green's-function theory to derive a set of self-consistent equations for the Green's function, self-energy, and polarization. As one of us has treated this derivation in detail in a previous paper,[12] it will merely be summarized here. In order to derive an expression for the dielectric response function, we consider a small external potential ϕ acting on a crystal containing N electrons. Then in the linear approximation the Hamiltonian is given by

$$H=\sum_i h(x_i)+\frac{1}{2}\sum_{i\neq j} v(\vec{r}_i,\vec{r}_j)+\sum_i \phi(x_i t)+V_{\text{nuclear}}, \tag{2.1}$$

where x includes space and spin coordinates $(x) = (\vec{r},\xi)$, h is the kinetic energy plus the interaction of the electron with the nuclei, v is the bare Coulomb interaction given by

$$v(\vec{r}_i,\vec{r}_j)=e^2/|\vec{r}_i-\vec{r}_j|, \tag{2.2}$$

and V_{nuclear} is the Coulomb repulsion of the nuclei which is treated as a constant here. After the final equations have been obtained, the external potential ϕ will be set equal to zero.

Using the field operator $\psi(x)$, the Hamiltonian can be written in second quantized form as follows:

$$\begin{aligned}H(t)=&\int \psi^\dagger(x)h(x)\psi(x)\,dx\\&+\frac{1}{2}\int \psi^\dagger(x)\psi^\dagger(x')v(\vec{r},\vec{r}\,')\psi(x')\psi(x)\,dx\,dx'\\&+\int \psi^\dagger(x)\psi(x)\phi(xt)\,dx+V_{\text{nuclear}},\end{aligned} \tag{2.3}$$

$$H(t)=H_0+H_1(t), \tag{2.4}$$

$$H_1(t)=\int \rho(x)\phi(xt)\,dx. \tag{2.5}$$

$$\rho(x)=\psi^\dagger(x)\psi(x). \tag{2.6}$$

Let $|t\rangle$ be the state of the N-particle system at time t. Then the time-evolution operator U is defined by

$$|t\rangle=U(tt')\,|t'\rangle. \tag{2.7}$$

It can be shown that the time-evolution operator satisfies the equation

$$U(t_1t_2)=U_0(t_1t_2)-i\int_{t_2}^{t_1} U_0(t_1t_3)H_1(t_3)U(t_3t_2)\,dt_3, \tag{2.8}$$

where U_0 is the time-evolution operator corresponding to H_0. Taking the functional derivative of both sides of (2.8) with respect to the external potential, one can obtain the following equation:

$$\frac{\delta U(t_1t_2)}{\delta\phi(x_3t_3)}=\begin{cases}-iU(t_1t_3)\,\rho(x_3)U(t_3t_2), & t_1>t_3>t_2\\ 0, & t_3>t_1, t_3<t_2.\end{cases} \tag{2.9}$$

Then defining the ground-state expectation value of a time-ordered product of Heisenberg operators as

$$\langle T[O_1(t_1)O_2(t_2)\cdots]\rangle=\frac{\langle N|U_0(t_0,-t_0)U(-t_0,t_0)T[O_1(t_1)O_2(t_2)\cdots]|N\rangle}{\langle N|U_0(t_0,-t_0)U(-t_0,t_0)|N\rangle} \tag{2.10}$$

and using (2.9), we have

$$\frac{\delta\langle T[O_1(t_1)O_2(t_2)\cdots]\rangle}{\delta\phi(xt)} = -i\langle T[\rho(xt)O_1(t_1)O_2(t_2)\cdots]\rangle + i\langle\rho(xt)\rangle\langle T[O_1(t_1)O_2(t_2)\cdots]\rangle , \tag{2.11}$$

$$\rho(xt) = \psi^\dagger(xt)\psi(xt) , \tag{2.12}$$

$$\psi(xt) = U(t_0t)\psi(x)U(tt_0) , \tag{2.13}$$

where $|N\rangle$ is the Heisenberg ground state of the N-particle system, O is a Heisenberg operator, T is the Dyson time-ordering operator, and $\psi(xt)$ is the Heisenberg field operator which satisfies the Heisenberg equation of motion

$$i\frac{\partial}{\partial t}\psi(xt) = [\psi(xt), H(t)] . \tag{2.14}$$

The one-electron Green's function is defined as

$$G(xt, x't') = -i\langle T[\psi(xt)\psi^\dagger(x't')]\rangle . \tag{2.15}$$

Then using Eqs. (2.3), (2.11), (2.14), and (2.15), one can show that the Green's function satisfies the following equation:

$$\left(i\frac{\partial}{\partial t_1} - h(x_1) - V(1)\right)G(12) - i\int v(1^+3)\frac{\delta G(12)}{\delta\phi(3)}\,d(3) = \delta(12) , \tag{2.16}$$

$$V(1) = \phi(1) + \int v(13)\langle\rho(3)\rangle\,d(3) , \tag{2.17}$$

where we have used the notation

$$(1) = (x_1, t_1) ,$$

$$(1^+) = (x_1, t_1 + \eta) ,$$

$$v(12) = v(\vec{r}_1, \vec{r}_2)\delta(t_1, t_2) ,$$

$$\delta(12) = \delta(x_1, x_2)\delta(t_1, t_2) ,$$

and $\lim_{\eta\to 0}$ is understood.

We define the screened interaction W, dielectric response function ϵ, and irreducible polarization propagator χ as follows:

$$W(12) = \int v(13)\epsilon^{-1}(32)\,d(3) , \tag{2.18}$$

$$\epsilon(12) = \delta(12) - \int \chi(13)v(32)\,d(3) , \tag{2.19}$$

$$\chi(12) = -i\frac{\delta G(22^+)}{\delta V(1)} . \tag{2.20}$$

Then defining the self-energy Σ as

$$\Sigma(12) = -i\int W(1^+3)G(14)\frac{\delta G^{-1}(42)}{\delta V(3)}\,d(34) , \tag{2.21}$$

Eq. (2.16) becomes

$$\left(i\frac{\partial}{\partial t_1} - h(x_1) - V(1)\right)G(12) - \int \Sigma(13)G(32)\,d(3) = \delta(12) . \tag{2.22}$$

Defining the vertex function Γ by

$$\Gamma(12,3) = -\frac{\delta G^{-1}(12)}{\delta V(3)} , \tag{2.23}$$

we can write the self-energy and polarization as

$$\Sigma(12) = i\int W(1^+3)G(14)\Gamma(42,3)d(34) , \tag{2.24}$$

$$\chi(12) = -i\int G(23)\Gamma(34,1)G(42^+)d(34) , \tag{2.25}$$

while the vertex function itself can be shown to satisfy

$$\Gamma(12,3) = \delta(12)\delta(13) + \int \frac{\delta\Sigma(12)}{\delta G(45)}G(46)\Gamma(67,3)G(75)\,d(4567) . \tag{2.26}$$

In order to complete the set of self-consistent equations, we combine (2.18) and (2.19) to obtain the following equation for the screened interaction:

$$W(12) = v(12) + \int v(13)\,\chi(34)\,W(42)d(34) . \tag{2.27}$$

The last four equations together with Eq. (2.22) can be iterated to obtain a whole hierarchy of approximations for Σ and χ.

Now that the final equations have been obtained, we set the external potential equal to zero in which case the dielectric function becomes a linear response function and the ground-state expectation value takes the usual form

$$\langle T[O_1(t_1)O_2(t_2)\cdots]\rangle = \langle N|\,T[O_1(t_1)O_2(t_2)\cdots]\,|N\rangle .$$

When the Hamiltonian for the system is independent of time, it is easily seen that the Green's function depends only on the difference of the time arguments. Then taking the Fourier transforms of (2.15) and (2.22) with respect to time, we can write the Green's function as

$$G(x, x', \omega) = \sum_k \frac{u_k(x)u_k^*(x')}{\omega - \epsilon_k} , \tag{2.28}$$

where ω is the frequency and the amplitudes u_k and energies ϵ_k satisfy the equation

$$[h(x) + V(x)]u_k(x) + \int \Sigma(x, x', \omega)u_k(x')\,dx' = \epsilon_k u_k(x) , \tag{2.29}$$

$$V(x) = \int v(\vec{r}, \vec{r}\,')\,\rho(x')\,dx' , \tag{2.30}$$

$$\rho(x)=\sum_{k\ \mathrm{occ}} u_k^*(x)u_k(x)\,. \tag{2.31}$$

If the self-energy Σ is independent of frequency, then the amplitudes and energies given by Eq. (2.29) are one-electron wave functions and energies and one can work in the one-electron energy band picture. Therefore in working with Eq. (2.29) we will always choose expressions for Σ that are independent of frequency.

III. RANDOM-PHASE APPROXIMATION

Equations (2.24)–(2.29) can be iterated to obtain successively more accurate approximations for the polarization or susceptibility χ. We start the iterations by setting Σ equal to zero

$$\Sigma_0=0\,. \tag{3.1}$$

Then Eq. (2.29) becomes the Hartree equation

$$[h(x)+V(x)]u_k(x)=\epsilon_k u_k(x) \tag{3.2}$$

while the vertex function, screened interaction, and susceptibility are given by

$$\Gamma_0(12,3)=\delta(12)\delta(13)\,, \tag{3.3}$$

$$W_0(12)=v(12)+\int v(13)\chi_0(34)W_0(42)\,d(34)\,, \tag{3.4}$$

$$\chi_0(12)=-iG(12^+)G(21)\,. \tag{3.5}$$

The above formula for the susceptibility is a well-known result called the random-phase approximation (RPA).

Taking the Fourier transform of (3.5) with respect to time, we have

$$\chi_{\mathrm{RPA}}(x_1,x_2,\omega)=-\frac{i}{2\pi}\int G(x_1,x_2,\omega')\times G(x_1,x_2,\omega'-\omega)\,d\omega'\,. \tag{3.6}$$

Then carrying out the ω' integration and taking the Fourier transform with respect to spatial coordinates, we obtain

$$\begin{aligned}\chi_{\mathrm{RPA}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega) &= -\frac{2}{V}(2\pi)^3\sum_{\vec{k}nn'}\frac{f_{\vec{k}+\vec{q}+\vec{K},n'}-f_{\vec{k}n}}{\epsilon_{\vec{k}+\vec{q}+\vec{K},n'}-\epsilon_{\vec{k}n}-\omega}\\ &\quad\times\langle\vec{k}n|e^{-i(\vec{q}+\vec{K}_1)\cdot\vec{r}}|\vec{k}+\vec{q}+\vec{K},n'\rangle\\ &\quad\times\langle\vec{k}n|e^{-i(\vec{q}+\vec{K}_2)\cdot\vec{r}}|\vec{k}+\vec{q}+\vec{K},n'\rangle^*\,,\end{aligned} \tag{3.7}$$

where $\vec{q}$ is a wave vector restricted to the first Brillouin zone and $\vec{K}_1$ and $\vec{K}_2$ are reciprocal lattice vectors. In the above expression, we have replaced the one-electron index k by a wave vector $\vec{k}$ restricted to the first Brillouin zone and a band index n; $\vec{K}$ is the reciprocal lattice vector such that $\vec{k}+\vec{q}+\vec{K}$ lies in the first zone; $f_{\vec{k}n}$ is the occupation number of the state $|\vec{k}n\rangle$; and V is the volume of the crystal. Having determined χ_{RPA}, we can then use (2.19) to obtain the RPA formula for the dielectric function

$$\epsilon_{\mathrm{RPA}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)=\delta_{\vec{K}_1,\vec{K}_2}-v(\vec{q})\chi_{\mathrm{RPA}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)\,, \tag{3.8}$$

$$v(\vec{q})=4\pi e^2/q^2\,. \tag{3.9}$$

As shown by Eq. (3.2), the wave functions and energies that appear in the RPA formula for the dielectric function are Hartree wave functions and energies. Therefore, in order to be consistent with the theory, one should use Hartree wave functions and energies to compute RPA dielectric functions.

IV. TIME-DEPENDENT HARTREE-FOCK APPROXIMATION

Since the RPA dielectric function corresponds to the Hartree equation, it contains no exchange terms. In order to include such terms in the dielectric function, we use Γ_0 to compute a higher-order expression for the self-energy

$$\Sigma_1(12)=iW_0(1^+2)G(12)\,. \tag{4.1}$$

This expression for Σ_1 is the first term of the expansion of Σ in powers of the screened interaction W. The screened interaction itself can then be expanded in powers of the bare instantaneous Coulomb interaction

$$W=v+v\chi v+\cdots\,. \tag{4.2}$$

If we keep the first term in this expansion, then W_0 becomes

$$\begin{aligned}W_0(1^+2)&=v(1^+2)\\ &=v(\vec{r}_1,\vec{r}_2)\delta(t_1-t_2+\eta)\end{aligned} \tag{4.3}$$

and the corresponding expression for Σ_1 is the Hartree-Fock approximation for the self-energy

$$\Sigma_{\mathrm{HF}}(x,x')=-\rho(x,x')v(\vec{r},\vec{r}')\,, \tag{4.4}$$

where ρ is the density matrix

$$\rho(x,x')=\sum_{k\ \mathrm{occ}} u_k^*(x')u_k(x)\,. \tag{4.5}$$

Putting Σ_{HF}, which is independent of frequency, into Eq. (2.29), we obtain the Hartree-Fock equation

$$[h(x)+V(x)]u_k(x)+\int \Sigma_{\mathrm{HF}}(x,x')u_k(x')\,dx'=\epsilon_k u_k(x)\,. \tag{4.6}$$

The expression for Σ_1 given in Eq. (4.1) leads to new expressions for the vertex function and susceptibility:

$$\Gamma_1(12,3)=\delta(12)\delta(13)+iW_0(1^+2)G(13)G(32)\,, \tag{4.7}$$

$$\chi_1(12)=-iG(12^+)G(21)+\int G(23)G(42+)W_0(3^+4)G(31)G(14)\,d(34)\,, \tag{4.8}$$

where W_0 is given by Eq. (4.3). Taking the Fourier transform of χ_1 with respect to time, we have

$$\chi_1(x_1,x_2,\omega)=-\frac{i}{2\pi}\int G(x_1,x_2,\omega')\,G(x_2,x_1,\omega'-\omega)\,d\omega'$$
$$-\frac{i^2}{(2\pi)^2}\int G(x_2,x_3,\omega'-\omega)\,G(x_4,x_2,\omega')W_0(\vec{r}_3,\vec{r}_4)\,G(x_3,x_1,\omega''-\omega)\,G(x_1,x_4,\omega'')\,d\omega'\,d\omega''\,. \tag{4.9}$$

Then integrating over ω' and ω'' and taking the Fourier transform with respect to spatial coordinates, we obtain the time-dependent Hartree-Fock (TDHF) formula for the susceptibility

$$\chi_{\mathrm{TDHF}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)=\chi_{\mathrm{RPA}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)$$
$$+\frac{4}{V^2}(2\pi)^3\sum_{\vec{k}\vec{k}'\vec{K}_3nn'mm'}\frac{\epsilon_{\vec{k}+\vec{q}+\vec{K},n'}-f_{\vec{k}n}}{\epsilon_{\vec{k}+\vec{q}+\vec{K},n'}-\epsilon_{\vec{k}n}-\omega}\;\frac{f_{\vec{k}'+\vec{q}+\vec{K}',m'}-f_{\vec{k}'m}}{\epsilon_{\vec{k}'+\vec{q}+\vec{K}',m'}-\epsilon_{\vec{k}'m}-\omega}$$
$$\times v(\vec{k}'-\vec{k}+\vec{K}_3)\langle\vec{k}n|e^{-i(\vec{q}+\vec{K}_1)\cdot\vec{r}}|\vec{k}+\vec{q}+\vec{K},n'\rangle\langle\vec{k}'m|e^{-i(\vec{q}+\vec{K}_2)\cdot\vec{r}}|\vec{k}'+\vec{q}+\vec{K}',m'\rangle^*$$
$$\times\langle\vec{k}+\vec{q}+\vec{K},n'|e^{-i(\vec{k}'-\vec{k}+\vec{K}_3)\cdot\vec{r}}|\vec{k}'+\vec{q}+\vec{K}',m'\rangle\langle\vec{k}n|e^{-i(\vec{k}'-\vec{k}+\vec{K}_3)\cdot\vec{r}}|\vec{k}'m\rangle^* \tag{4.10}$$

which leads to the time-dependent Hartree-Fock formula for the dielectric function

$$\epsilon_{\mathrm{TDHF}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)=\delta_{\vec{K}_1,\vec{K}_2}-v(\vec{q})\chi_{\mathrm{TDHF}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)\,, \tag{4.11}$$

$$\epsilon_{\mathrm{TDHF}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)=\epsilon_{\mathrm{RPA}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)-v(\vec{q})\chi_{\mathrm{ex}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)\,, \tag{4.12}$$

$$\chi_{\mathrm{ex}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)=\chi_{\mathrm{TDHF}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)-\chi_{\mathrm{RPA}}(\vec{q}+\vec{K}_1,\vec{q}+\vec{K}_2,\omega)\,. \tag{4.13}$$

As shown by Eq. (4.12), the first term in the TDHF dielectric function is the RPA dielectric function. According to Eq. (4.6), the wave functions and energies that appear in all of the terms in ϵ_{TDHF}, including the first term ϵ_{RPA}, are Hartree-Fock wave functions and energies. Therefore in order to be consistent with the theory, one should use Hartree-Fock wave functions and energies to compute TDHF dielectric functions.

V. NUMERICAL RESULTS

In order to illustrate the importance of choosing the proper wave functions and energies to use in a particular formula for the dielectric function or susceptibility, we have numerically computed the RPA susceptibility for the free-electron gas. In

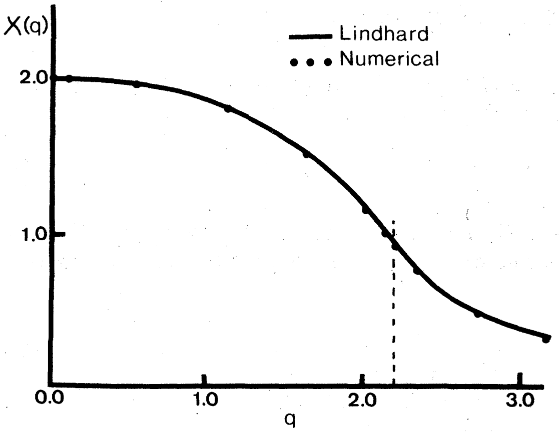

FIG. 1. χ_{RPA} computed numerically with Hartree energies, and the Lindhard susceptibility. Both χ_{RPA} and the Lindhard susceptibility are normalized to 2 at $\vec{q}=0$ and are computed for $\omega=0$ at points along the Δ axis. The units of $\vec{q}$ are chosen so that $\vec{q}=2$ at the point H. The dotted vertical line corresponds to $q=2k_F$, where k_F is the Fermi wave vector.

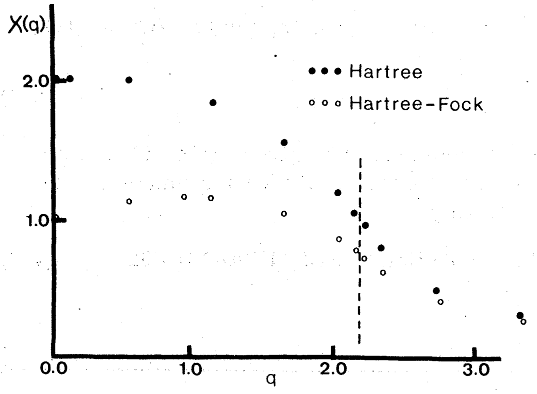

FIG. 2. χ_{RPA} computed numerically with both Hartree and Hartree-Fock energies. The formula for χ_{RPA} is normalized so that it is equal to 2 at $\vec{q}=0$ when Hartree energies are used. The susceptibilities are computed for $\omega=0$ at points along the Δ axis, where the units of $\vec{q}$ agree with Fig. 1.

this system the exact answer is known and is given by the Lindhard formula[13] when Hartree energies are used in the calculation. This provides an important check for our computer programs which are quite complex due to the principal values which must be taken in the expression for χ_{RPA}. For crystals with band gaps such as insulators and semiconductors the energy denominator in (3.7) is never zero and the sum over $\vec{k}$ can be done by simple numerical sampling of the Brillouin zone; however, in the case of metals or the free-electron gas, there is no energy gap between the filled and empty states and hence the sum over $\vec{k}$ must be converted to an integral so that principal values can be taken. In order to evaluate these principal-value integrals, we have employed the analytic tetrahedron method[14] and have written the required computer programs for this method which may be applied to crystals with arbitrary energy band structures.

As a test of our programs, we have computed the RPA susceptibility for the electron gas using Hartree energies, which are the ones that are called for in the RPA formula. In Fig. 1 the results are shown and are compared with the exact answer as given by the Lindhard formula. The programs were set up for a body-centered cubic Brillouin zone which was divided up into tetrahedrons. In order to apply our programs to the free-electron gas, we choose a Fermi wave vector equal to 0.55 times the distance from Γ to H in which case the Fermi sphere and the points $\vec{k}+\vec{q}$ for all values of $\vec{q}$ considered lie within the Brillouin zone. The calculation was done for the diagonal part of χ_{RPA} using a division of 24 points along the Δ axis which corresponds to 3456 tetrahedrons in $\frac{1}{48}$ of the Brillouin zone. As shown by Fig. 1, the results are in excellent agreement with the Lindhard formula, indicating that our analytic tetrahedron method programs are working properly.

We then computed the RPA susceptibility for the free-electron gas using Hartree-Fock energies, which are inconsistent with the RPA formula. The required integrals are too complicated to evaluate directly as Lindhard did, but the analytical tetrahedron method was easily applied. The results differ substantially from the RPA susceptibility computed with Hartree energies, as shown in Fig. 2. Thus different types of energies lead to substantially different results for χ_{RPA}, and therefore it is very important to use the proper type of energies and wave functions when one is computing RPA susceptibilities. As clearly shown in Sec. III, the wave functions and energies that are consistent with the RPA formula are Hartree wave functions and energies.

Although χ_{RPA} computed with Hartree-Fock energies is inconsistent with the theory when considered by itself, it does have a proper use as it is the first term in the TDHF susceptibility, which is given by Eq. (4.10). The full TDHF susceptibility calculation will be presented in a later paper, in which the convergence of the susceptibility function with respect to the successive approximations to Σ will be examined.

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