Bilinear Electromagnetic Response of an Electron Gas in Many-Body Perturbation Theory

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The bilinear response of an interacting electron gas to an external electromagnetic field is investigated by many-body perturbation theory. The Coulomb interaction and the interaction of the electrons with the external field are treated as perturbations on the motion of the free electrons. The analysis is based on the pair approximation introduced by Gell-Mann and Brueckner to treat the Coulomb interaction up to all orders. In the perturbation expansion, terms up to the second order in the external field are retained in order to calculate expectation values of the current and charge densities in the perturbed state. By interpreting the results obtained for the linear and bilinear current densities in terms of the induced field set up in the high-density electron gas, these are shown to be equivalent to the results which may be derived by using a self-consistent-field approach.

I. INTRODUCTION

FOR a weak external probe it is enough to consider only the linear response of a physical system. However, when one deals with the interaction of the system with a sufficiently strong external field, one has to go beyond this linear theory. For finding the bilinear electromagnetic response of an electron gas, one has to calculate the part of the induced polarization $\mathbf{P}$ or the current density $\mathbf{J}=\partial\mathbf{P}/\partial t$, which varies bilinearly with the external field. Recently, a self-consistent-field (SCF) technique to calculate the induced nonlinear polarization of an electron gas has been developed.[1-4] The external electromagnetic field is assumed to contain both the longitudinal and transverse components. In this approach one considers the electrons to be moving independently in a SCF which is set up due to the combined effect of the induced motion of the electrons and the external field. This field is the self-consistent solution of Maxwell's equations and the equation describing the motion of the electrons. One does not consider the Coulomb interactions among the electrons explicitly in the SCF calculation. Ehrenreich and Cohen[5] have, however, shown that the SCF result for the linear longitudinal polarization of an electron gas is equivalent to that calculated in the "random phase approximation" developed by Bohm and Pines[6] for treating the electron interactions. This result may also be obtained in a similar approximation called the "pair approximation", introduced by Gell-Mann and Brueckner[7,8] to treat the Coulomb interaction up to all orders in diagrammatic many-body perturbation theory. The important feature of the pair approximation is that the energy-momentum transfer at each Coulomb vertex in any diagram is assumed to be always the same. For a high-density electron gas, these diagrams give the most dominant contribution.

We consider our physical system to be a gas of electrons with a uniform background of static positive charge equal and opposite to the average charge density of the electrons. We treat the Coulomb interaction among the electrons together with the external interaction as perturbations on the motion of free electrons. We then explicitly calculate both the linear and bilinear induced current densities in the electron gas. The external field is supposed to be either longitudinal or transverse and the Coulomb interaction is treated in the pair approximation. We work in a gauge where the external scalar potential is zero and the electromagnetic fields $\mathbf{E}$ and $\mathbf{H}$ are expressed in terms of a vector potential $\mathbf{A}$ alone. This means that in our approach the interaction of the electrons and the external field is momentum-dependent even for a longitudinal probe. To simplify the algebra we assume the external vector potential $\mathbf{A}$ to be represented by a plane wave varying with a single frequency ω. In this case the bilinear induced current density has components varying with frequencies zero and 2ω. We calculate in this paper only the part which varies with frequency 2ω. In order to obtain bilinear current density for the case where the external field contains more than one Fourier component, it should be emphasized at this point that it is not enough to obtain the result for a single frequency component and sum this over different Fourier components. This is possible only in the case of the linear response theory. However, the extension of our calculation to the general case is not very difficult and we would restrict ourselves to a single Fourier component in the external potential.

In Sec. II we describe the mathematical formulation of the problem. In Sec. III we sum up the contributions to the induced current density from all those terms in the perturbation expansion which are linear in the external field. In Sec. IV we sum up all the diagrams up to

[1] A. S. Pine, Phys. Rev. **139**, A901 (1965).
[2] S. S. Jha, Phys. Rev. **145**, 500 (1966).
[3] For the classical SCF approach see S. S. Jha, Phys. Rev. **140**, A2020 (1965).
[4] N. Bloembergen and Y. R. Shen, Phys. Rev. **141**, 298 (1966).
[5] H. Ehrenreich and M. H. Cohen, Phys. Rev. **115**, 786 (1959).
[6] D. Bohm and D. Pines, Phys. Rev. **92**, 609 (1953); P. Nozières and D. Pines, Nuovo Cimento **9**, 470 (1958).
[7] M. Gell-Mann and K. A. Brueckner, Phys. Rev. **106**, 364 (1957); J. Hubbard, Proc. Roy. Soc. (London) **A243**, 336 (1957); A. J. Glick, Ann. Phys. (N. Y.) **17**, 61 (1962).
[8] C. Kittel, *Quantum Theory of Solids* (John Wiley & Sons, Inc., New York, 1963), Chap. 6.

second order in the external field and thus find the induced polarization which varies bilinearly with the external field. The coefficient of the bilinear polarization is the bilinear response function for the electron gas. A careful interpretation of our results shows that these are equivalent to expressions which one would obtain if one used the SCF technique for his problem.

II. MATHEMATICAL FORMULATION

Let us consider an electron gas in the presence of an external vector potential $\mathbf{A}(\mathbf{x},t)$ given by[9]

$$\mathbf{A}(\mathbf{x},t)=\mathbf{a}_1 e^{i\mathbf{q}\cdot\mathbf{x}}e^{-i\omega t}, \tag{2.1}$$

where $\mathbf{a}_1$ is either longitudinal or transverse and where ω is assumed to contain an arbitrarily small positive imaginary part. The Hamiltonian for such a system may be written as

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}', \tag{2.2}$$

where $\mathcal{H}_0$ is the kinetic energy of the electrons and the perturbation $\mathcal{H}'$ contains their Coulomb interaction and the interaction with the external field. In the notation of the second-quantization formalism,[10] these are given by

$$\mathcal{H}_0=\sum_{\mathbf{k}}E_{\mathbf{k}}C_{\mathbf{k}}^{\dagger}C_{\mathbf{k}}, \tag{2.3}$$

$$E_{\mathbf{k}}=\hbar^2k^2/2m, \tag{2.4}$$

and

$$\mathcal{H}'=\mathcal{H}_c+\mathcal{H}_{ex}, \tag{2.5}$$

where

$$\mathcal{H}_c=\tfrac{1}{2}\sum_{\mathbf{q}'}{}'\sum_{\mathbf{k}'}\sum_{\mathbf{k}''}V(q')\times[C_{\mathbf{k}'+\mathbf{q}'}^{\dagger}C_{\mathbf{k}'}C_{\mathbf{k}''}^{\dagger}C_{\mathbf{k}''+\mathbf{q}'}-n], \tag{2.6}$$

$$V(q')=4\pi e^2/q'^2, \tag{2.7}$$

$$\mathcal{H}_{ex}=h_1+h_2, \tag{2.8}$$

$$h_1=\sum_{\mathbf{k}}\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle C_{\mathbf{k}+\mathbf{q}}^{\dagger}C_{\mathbf{k}}e^{-i\omega t}, \tag{2.9}$$

$$h_2=\sum_{\mathbf{k}}\langle\mathbf{k}+2\mathbf{q}|v_2|\mathbf{k}\rangle C_{\mathbf{k}+2\mathbf{q}}^{\dagger}C_{\mathbf{k}}e^{-2i\omega t}, \tag{2.10}$$

$$\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle=(e\hbar/mc)\mathbf{a}_1\cdot(\mathbf{k}+\tfrac{1}{2}\mathbf{q}), \tag{2.11}$$

$$\langle\mathbf{k}+2\mathbf{q}|v_2|\mathbf{k}\rangle=(e^2/2mc^2)\mathbf{a}_1\cdot\mathbf{a}_1. \tag{2.12}$$

In Eq. (2.6) the summation over $\mathbf{q}'$ excludes the term $\mathbf{q}'=0$ and n is the number density of the electrons.

At time $t=-\infty$, let the system be in the unperturbed ground state Φ_0 (single-particle momentum states filled up to the Fermi momentum $\hbar k_f$). In the interaction representation the state of the system at time t may then be written as

$$\Phi(t)=U(t,-\infty)\Phi_0, \tag{2.13}$$

where

$$U(t,-\infty)=1+\sum_{n=1}^{\infty}\left(\frac{-i}{\hbar}\right)^n\int_{-\infty}^{t}dt_1\int_{-\infty}^{t_1}dt_2\cdots\int_{-\infty}^{t_{n-1}}dt_n\{\mathcal{H}'(t_1)\mathcal{H}'(t_2)\cdots\mathcal{H}'(t_n)\}, \tag{2.14}$$

and

$$\mathcal{H}'(t)=e^{(i/\hbar)\mathcal{H}_0t}\mathcal{H}'e^{(-i/\hbar)\mathcal{H}_0t}=\mathcal{H}_c(t)+h_1(t)+h_2(t), \tag{2.15}$$

$$\mathcal{H}_c(t)=\tfrac{1}{2}\sum_{\mathbf{q}'}{}'\sum_{\mathbf{k}'}\sum_{\mathbf{k}''}V(q')[C_{\mathbf{k}'+\mathbf{q}'}^{\dagger}C_{\mathbf{k}'}C_{\mathbf{k}''}^{\dagger}C_{\mathbf{k}''+\mathbf{q}'}e^{(i/\hbar)(E_{\mathbf{k}'+\mathbf{q}'}-E_{\mathbf{k}'}-E_{\mathbf{k}''+\mathbf{q}'}+E_{\mathbf{k}''})t}-n], \tag{2.16}$$

$$h_1(t)=\sum_{\mathbf{k}}\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle C_{\mathbf{k}+\mathbf{q}}^{\dagger}C_{\mathbf{k}}e^{(i/\hbar)(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega)t}, \tag{2.17}$$

$$h_2(t)=\sum_{\mathbf{k}}\langle\mathbf{k}+2\mathbf{q}|v_2|\mathbf{k}\rangle C_{\mathbf{k}+2\mathbf{q}}^{\dagger}C_{\mathbf{k}}e^{(i/\hbar)(E_{\mathbf{k}+2\mathbf{q}}-E_{\mathbf{k}}-2\hbar\omega)t}. \tag{2.18}$$

Since we are interested in only those terms in U which are either linear or bilinear in $\mathbf{a}_1$, let us write

$$U=1+U_1+U_2+\cdots, \tag{2.19}$$

$$U^{-1}=1-U_1-U_2+U_1U_1+\cdots, \tag{2.20}$$

where U_1 and U_2 contain terms which are linear and bilinear, respectively, in the external potential. In the interaction representation the operators for the charge density ρ and the current density $\mathbf{J}$ are

$$\rho_{op}(t)=-e\sum_{\mathbf{q}'}e^{i\mathbf{q}'\cdot\mathbf{x}}\sum_{\mathbf{k}}C_{\mathbf{k}}^{\dagger}C_{\mathbf{k}+\mathbf{q}'}e^{(-i/\hbar)(E_{\mathbf{k}+\mathbf{q}'}-E_{\mathbf{k}})t} \tag{2.21}$$

[9] Strictly speaking, we should add the complex conjugate term corresponding to the first term on the right of Eq. (2.1). However, we are not interested in calculating the bilinear polarization varying with frequency zero. The part of the bilinear polarization varying as $e^{2i\omega t}$ is the quantity which is the complex conjugate to the part of the bilinear polarization varying as $e^{-2i\omega t}$, which we would calculate in this paper.

[10] See Ref. 8.

and

$$\mathbf{J}_{\rm op}(t)=-\frac{eh}{m}\sum_{\mathbf{q}'}e^{i\mathbf{q}'\cdot\mathbf{x}}\sum_{\mathbf{k}}(\mathbf{k}+\tfrac{1}{2}\mathbf{q}')C_{\mathbf{k}}^{\dagger}C_{\mathbf{k}+\mathbf{q}'}e^{(i/\hbar)(E_{\mathbf{k}+\mathbf{q}'}-E_{\mathbf{k}})t}-\frac{e^2}{mc}\mathbf{a}_1\sum_{\mathbf{q}'}e^{i\mathbf{q}'\cdot\mathbf{x}}\sum_{\mathbf{k}}C_{\mathbf{k}}^{\dagger}C_{\mathbf{k}+\mathbf{q}'-\mathbf{q}}e^{(-i/\hbar(E_{\mathbf{k}+\mathbf{q}'-\mathbf{q}}-E_{\mathbf{k}})t}, \quad (2.22)$$

respectively.

The induced linear charge and current densities vary as $e^{i\mathbf{q}\cdot\mathbf{x}}e^{-i\omega t}$ because of the form of the external potential given by Eq. (2.1). For the same reason the bilinear induced current density varies as $e^{2i\mathbf{q}\cdot\mathbf{x}}e^{-2i\omega t}$, and it may also be shown[11] to be longitudinal if $\mathbf{a}_1$ is either longitudinal or transverse. The longitudinal part of the current density and the polarization may be obtained from the knowledge of the induced charge density alone, by using the relation

$$\partial\rho/\partial t=-\nabla\cdot\partial\mathbf{P}/\partial t=-\nabla\cdot\mathbf{J}. \quad (2.23)$$

Thus in the present case, it is enough to know the current density $J_1(\mathbf{q})e^{i\mathbf{q}\cdot\mathbf{x}}e^{-i\omega t}$, which is of the first order in the external potential, and the charge density $\rho_2(2\mathbf{q})e^{2i\mathbf{q}\cdot\mathbf{x}}e^{-2i\omega t}$ which is of the second order in the external potential. The expectation value of any operator $O_{\rm op}$ at time t is given by

$$\langle O\rangle=\langle\Phi_0|\,U^{-1}O_{\rm op}(t)U|\,\Phi_0\rangle. \quad (2.24)$$

From Eqs. (2.19)–(2.23) we therefore find

$$\mathbf{J}_1(\mathbf{q})=-(e^2\mathbf{a}_1/mc)\langle\Phi_0|\sum_{\mathbf{k}}C_{\mathbf{k}}^{\dagger}C_{\mathbf{k}}|\Phi_0\rangle-(eh/m)\langle\Phi_0|\,[\sum_{\mathbf{k}}(\mathbf{k}+\tfrac{1}{2}\mathbf{q})C_{\mathbf{k}}^{\dagger}C_{\mathbf{k}+\mathbf{q}}e^{(-i/\hbar)(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega)t},\,U_1]|\,\Phi_0\rangle, \quad (2.25)$$

and

$$\mathbf{J}_2(2\mathbf{q})=\frac{w\mathbf{q}}{q^2}\rho_2(2\mathbf{q})=-\frac{ew\mathbf{q}}{q^2}\langle\Phi_0|\,[\sum_{\mathbf{k}}C_{\mathbf{k}}^{\dagger}C_{\mathbf{k}+2\mathbf{q}}e^{(-i/\hbar)(E_{\mathbf{k}+2\mathbf{q}}-E_{\mathbf{k}}-2\hbar\omega)t},\,U_2]|\,\Phi_0\rangle$$
$$-\frac{ew\mathbf{q}}{q^2}\langle\Phi_0|\,[U_1\sum_{\mathbf{k}}C_{\mathbf{k}}^{\dagger}C_{\mathbf{k}+2\mathbf{q}}e^{(-i/\hbar)(E_{\mathbf{k}+2\mathbf{q}}-E_{\mathbf{k}}-2\hbar\omega)t},\,U_1]|\,\Phi_0\rangle. \quad (2.26)$$

In the next two section we will calculate U_1 and U_2 to find $\mathbf{J}_1(\mathbf{q})$ and $\mathbf{J}_2(\mathbf{q})$.

III. LINEAR INDUCED CURRENT DENSITY

For calculating the linear response to the external perturbation it is enough to know the operator U_1. U_1 consists of all the terms in Eq. (2.14) which are linear in h_1 and which do not involve h_2. The contribution of the term which involves no Coulomb interaction may easily be obtained after a simple integration, and it is given by

$$U_1^{(0)}=-\sum_{\mathbf{k}}\langle\mathbf{k}+q|\,v_1|\,\mathbf{k}\rangle\frac{e^{(i/\hbar)(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega)t}}{E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar w-i\epsilon}C_{\mathbf{k}+\mathbf{q}}^{\dagger}C_{\mathbf{k}}. \quad (3.1)$$

The contribution of the terms which involve the Coulomb interaction only once (see Fig. 1) may be calculated in the pair approxima tion and it may be written as

$$U_1^{(1)}=V(q)\sum_{\mathbf{k}'}\langle\mathbf{k}'+\mathbf{q}|\,v_1|\,\mathbf{k}'\rangle M(\mathbf{k}',\mathbf{q},w)\sum_{\mathbf{k}}\frac{e^{(i/\hbar)(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega)t}}{E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar w-i\epsilon}C_{\mathbf{k}+\mathbf{q}}^{\dagger}C_{\mathbf{k}}, \quad (3.2)$$

where by definition

$$M(\mathbf{k}',\mathbf{q},\omega)=-\frac{f_0(E_{\mathbf{k}'+\mathbf{q}})-f_0(E_{\mathbf{k}'})}{E_{\mathbf{k}'+\mathbf{q}}-E_{\mathbf{k}'}-\hbar w-i\epsilon}, \quad (3.3)$$

and where in anticipation of the fact that U_1 finally is going to operate on the ground state Φ_0, we have replaced

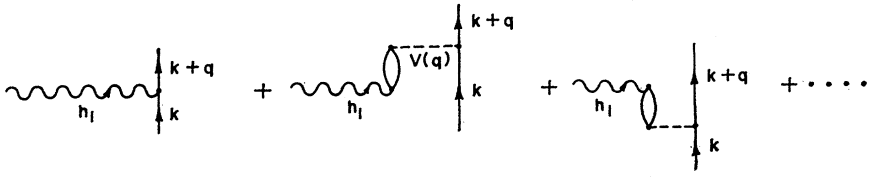

FIG. 1. Diagrams which are considered to calculate U_1.

[11] The bilinear terms which can occur are of the form $(\mathbf{a}_1\cdot\mathbf{q})\mathbf{a}_1$, $(\mathbf{a}_1\cdot\mathbf{a}_1)\mathbf{q}$ and $(\mathbf{a}_1\cdot\mathbf{q})^2\mathbf{q}$.

the operators like $C_{k'}^{\dagger}C_{k'}$ by $f_0(E_{k'})$, the Fermi distribution function. The problem of summing $U_1^{(0)}$, $U_1^{(1)}$, $U_1^{(2)}$ etc., may be transformed into an integral equation and the solution of this gives the sum U_1 as

$$U_1=U_1^{(0)}+U_1^{(1)}+U_1^{(2)}+\cdots=-\sum_{\mathbf{k}}\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle_c\frac{e^{(i/\hbar)(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega)t}}{E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar w-i\epsilon}C_{\mathbf{k}+\mathbf{q}}^{\dagger}C_{\mathbf{k}}, \tag{3.4}$$

where

$$\langle\mathbf{k}+\mathbf{q}|v_1|\rangle_c=\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle - V(q)\frac{\sum_{\mathbf{k}'}\langle\mathbf{k}'+\mathbf{q}|v_1|\mathbf{k}'\rangle M(\mathbf{k}',\mathbf{q},\omega)}{1+V(q)M(\mathbf{q},\omega)} \tag{3.5}$$

and where

$$M(\mathbf{q},\omega)=\sum_{\mathbf{k}}M(\mathbf{k},\mathbf{q},\omega)=-\sum_{\mathbf{k}}\frac{f_0(E_{\mathbf{k}+\mathbf{q}})-f_0(E_{\mathbf{k}})}{E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega-i\epsilon}. \tag{3.6}$$

If we note that $f_0(E_{\mathbf{k}})$ is an even function of $\mathbf{k}$ and that the $\mathbf{k}$-dependent term in $E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}$ is of the form $\mathbf{k}\cdot\mathbf{q}$, then by using Eq. (2.11) we may show that the second term on the right-hand side of Eq. (3.5) is zero if $\mathbf{a}_1$ is transverse. This implies the well-known fact that in our approximation there is no screening of the external potential if it is transverse. Let us, therefore, split $\mathbf{a}_1$ as

$$\mathbf{a}_1=(\mathbf{a}_1\cdot\mathbf{q}/q^2)\mathbf{q}+(\mathbf{q}\times\mathbf{a}_1)\times\mathbf{q}/q^2, \tag{3.7}$$

where the first term is zero if $\mathbf{a}_1$ is transverse and the second term is zero if it is longitudinal. Equation (3.5) may then be simplified to

$$\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle_c=\frac{e\hbar}{mc}\mathbf{a}_1\cdot(\mathbf{k}+\tfrac{1}{2}\mathbf{q}) - \frac{e\omega}{c}\frac{\mathbf{a}_1\cdot\mathbf{q}}{q^2}\frac{V(q)M(\mathbf{q},\omega)}{1+V(q)M(\mathbf{q},\omega)}. \tag{3.8}$$

Using Eqs. (2.25), (3.3), and (3.4) the linear current density may now be calculated and we find

$$\mathbf{J}_1(\mathbf{q})=-\frac{e^2n}{mc}\mathbf{a}_1+\frac{e\hbar}{m}\sum_{\mathbf{k}}(\mathbf{k}+\tfrac{1}{2}\mathbf{q}) \times\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle_c M(\mathbf{k},\mathbf{q},\omega). \tag{3.9}$$

Using Eqs. (3.7) and (3.8), this may also be split into longitudinal and transverse components and we may reduce them to the form

$$\mathbf{J}_1^{\text{long}}(\mathbf{q})=\frac{e^2\omega^2}{cq^2}\mathbf{a}_1^{\text{long}}\frac{M(\mathbf{q},\omega)}{1+V(q)M(\mathbf{q},\omega)}, \tag{3.10}$$

and

$$\mathbf{J}_1^{\text{tr}}(\mathbf{q})=-\frac{e^2n}{mc}\mathbf{a}_1^{\text{tr}}+\frac{e^2\hbar^2}{m^2c}\mathbf{a}_1^{\text{tr}}\sum_{k}\frac{(\mathbf{k}\times\mathbf{q})^2}{q^2}M(\mathbf{k},\mathbf{q},\omega). \tag{3.11}$$

In order to compare these with the SCF calculation let us now relate the external potential $\mathbf{a}_1$ to the SCF potential $\mathbf{a}_1(\text{SCF})$ by the relation $\mathbf{E}(\text{SCF})+4\pi\mathbf{P}=\mathbf{E}_{\text{ext}}$. With the use of Maxwell's equations this implies

$$(\mathbf{q}\times\mathbf{a}_1^{\text{tr}}(\text{SCF}))\times\mathbf{q}=\frac{\omega^2}{c^2}\mathbf{a}_1^{\text{tr}}(\text{SCF}) +\frac{4\pi}{c}\mathbf{J}_1^{\text{tr}}(q)=(\mathbf{q}\times\mathbf{a}_1^{\text{tr}})\times\mathbf{q}) \tag{3.12}$$

and

$$\mathbf{a}_1^{\text{long}}(\text{SCF})+(4\pi c/\omega^2)\mathbf{J}_1^{\text{long}}(q)=\mathbf{a}_1^{\text{long}}. \tag{3.13}$$

From Eqs. (3.10), (3.12), and (3.13) we therefore find

$$\mathbf{a}_1^{\text{tr}}(\text{SCF})=\mathbf{a}_1^{\text{tr}} \tag{3.14}$$

and

$$\mathbf{a}_1^{\text{long}}(\text{SCF})=\frac{\mathbf{a}_1^{\text{long}}}{1+V(q)M(\mathbf{q},\omega)}. \tag{3.15}$$

Substitutions of these relations in Eqs. (3.10) and (3.11) lead to the results which are identical with the SCF calculation.[12] The longitudinal dielectric constant $\epsilon(\mathbf{q},\omega)$ in our notation is just the quantity $1+V(q)M(\mathbf{q},\omega)$.

IV. BILINEAR INDUCED CURRENT DENSITY

In order to calculate the current density $\mathbf{J}_2(2\mathbf{q})$ from Eq. (2.26) we must first find U_2. U_2 contains all those terms in Eq. (2.14) which are either bilinear in h_1 or linear in h_2. The contribution of the diagrams with no Coulomb interaction (see Fig. 2) may be written as

$$U_2^{(0)}=\sum_{\mathbf{k}}\left\{\langle\mathbf{k}+2\mathbf{q}|v_1|\mathbf{k}+\mathbf{q}\rangle\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle\left[\frac{1-f_0(E_{\mathbf{k}+\mathbf{q}})}{E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega-i\epsilon}-\frac{f_0(E_{\mathbf{k}+\mathbf{q}})}{E_{\mathbf{k}+2\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega-i\epsilon}\right] -\frac{e^2}{2mc^2}\mathbf{a}_1\cdot\mathbf{a}_1\right\}\frac{e^{(i/\hbar)(E_{\mathbf{k}+2\mathbf{q}}-E_{\mathbf{k}}-2\hbar\omega)t}}{E_{\mathbf{k}+2\mathbf{q}}-E_{\mathbf{k}}-2\hbar\omega-2i\epsilon}C_{\mathbf{k}+2\mathbf{q}}^{\dagger}C_{\mathbf{k}}, \tag{4.1}$$

where again we have replaced operators of the form $C_{\mathbf{k}+\mathbf{q}}^{\dagger}C_{\mathbf{k}+\mathbf{q}}$ by $f_0(E_{\mathbf{k}+\mathbf{q}})$. One may introduce Coulomb inter-

[12] Compare with Eqs. (3.11) and (3.13)–(3.16) of Ref. 2, after changing the summation $(2\pi)^{-3}\sum_{\mathbf{k}}$ (occupied states) of that paper to $\sum_{\mathbf{k}}f_0(E_{\mathbf{k}})$. Also, $\mathbf{a}_1$ in that paper is our $\mathbf{a}_1(\text{SCF})$.

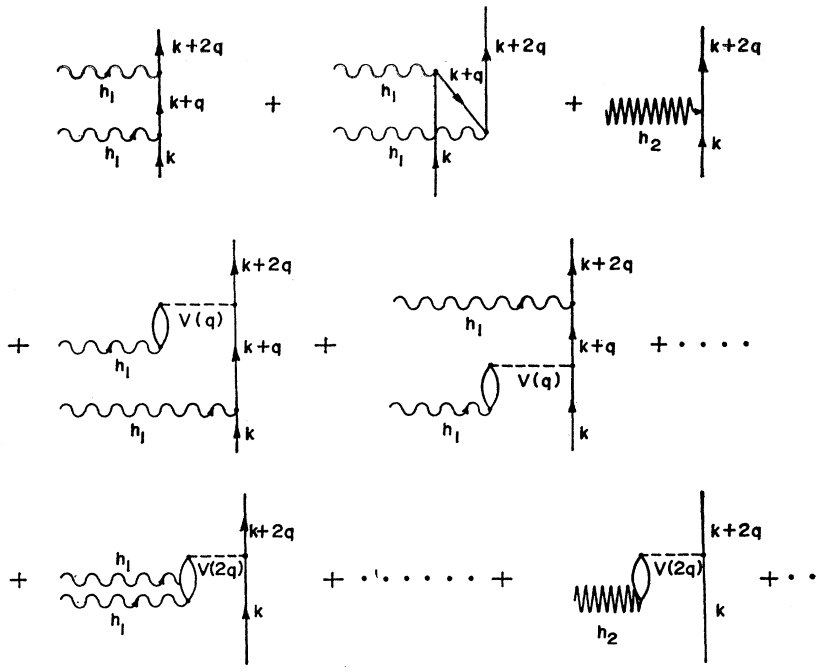

FIG. 2. Types of diagrams considered for U_2. All the diagrams with different time orderings have not been shown.

actions in the diagrams containing h_1 twice, in two different ways. We would either modify these two interaction vertices separately or let them act together and then introduce the modifications (see Fig. 2). When we sum up all such diagrams and diagrams containing h_2, we obtain

$$U_2=\sum_{\mathbf{k}}\Bigg\{\langle\mathbf{k}+2\mathbf{q}|v_1|\mathbf{k}+\mathbf{q}\rangle_c\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle_c\left[\frac{1-f_0(E_{\mathbf{k}+\mathbf{q}})}{E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega-i\epsilon}-\frac{f_0(E_{\mathbf{k}+\mathbf{q}})}{E_{\mathbf{k}+2\mathbf{q}}-E_{\mathbf{k}+\mathbf{q}}-\hbar\omega-i\epsilon}\right]$$
$$-\frac{V(2\mathbf{q})}{[1+V(2q)M(2\mathbf{q},2\omega)]}\sum_{\mathbf{k}'}\langle\mathbf{k}'+2\mathbf{q}|v_1|\mathbf{k}'+\mathbf{q}\rangle_c\langle\mathbf{k}'+\mathbf{q}|v_1|\mathbf{k}'\rangle_c N(\mathbf{k}',\mathbf{q},\mathbf{q},\omega,\omega)$$
$$-\frac{e^2}{2mc^2}\frac{\mathbf{a}_1\cdot\mathbf{a}_1}{[1+V(2q)M(2\mathbf{q},2\omega)]}\Bigg\}\frac{e^{(i/\hbar)(E_{\mathbf{k}+2\mathbf{q}}-E_{\mathbf{k}}-2\hbar\omega)t}}{E_{\mathbf{k}+2\mathbf{q}}-E_{\mathbf{k}}-2\hbar\omega-2i\epsilon}C_{\mathbf{k}+2\mathbf{q}}{}^{\dagger}C_{\mathbf{k}}\,, \quad (4.2)$$

where

$$N(\mathbf{k}',\mathbf{q},\mathbf{q},\omega,\omega)=\frac{1}{E_{\mathbf{k}'+2\mathbf{q}}-E_{\mathbf{k}'}-2\hbar\omega-2i\epsilon}\left\{\frac{f_0(E_{\mathbf{k}'+2\mathbf{q}})-f_0E_{\mathbf{k}'+\mathbf{q}})}{E_{\mathbf{k}'+2\mathbf{q}}-E_{\mathbf{k}'+\mathbf{q}}-\hbar\omega-i\epsilon}-\frac{f_0(E_{\mathbf{k}'+\mathbf{q}})-f_0(E_{\mathbf{k}'})}{E_{\mathbf{k}'+\mathbf{q}}-E_{\mathbf{k}'}-\hbar\omega-i\epsilon}\right\}. \quad (4.3)$$

The quantities $M(2\mathbf{q},2\omega)$, $\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle_c$, and $\langle\mathbf{k}+2\mathbf{q}|v_1|\mathbf{k}+\mathbf{q}\rangle_c$ needed in Eqs. (4.2) and (4.3) may be obtained from Eqs. (3.6) and (3.8).

From Eqs. (2.26), (3.4), (4.2), and (4.3) we then find

$$\mathbf{J}_2(2\mathbf{q})=-\frac{e\omega\mathbf{q}}{q^2\epsilon(2\mathbf{q},2\omega)}\sum_{\mathbf{k}}\langle\mathbf{k}+2\mathbf{q}|v_1|\mathbf{k}+\mathbf{q}\rangle_c\langle\mathbf{k}+\mathbf{q}|v_1|\mathbf{k}\rangle_c N(\mathbf{k},\mathbf{q},\mathbf{q},\omega,\omega)+\frac{\omega\mathbf{q}}{q^2\epsilon(2\mathbf{q},2\omega)}\frac{e^3}{2mc^2}\mathbf{a}_1\cdot\mathbf{a}_1M(2\mathbf{q},2\omega)\,, \quad (4.4)$$

where

$$\epsilon(2\mathbf{q},2\omega)=1+V(2q)M(2\mathbf{q},2\omega)\,. \quad (4.5)$$

We may rewrite the above results in terms of the self-consistent potentials $\mathbf{a}_1$(SCF) by using Eqs. (3.8), (3.14), and (3.15). These expressions may be simplified further by splitting $\mathbf{a}_1$(SCF) according to Eq. (3.7), and using the known properties of $f_0(E_{\mathbf{k}})$ and the known forms of energy denominators in M and N. With these simplifications we may reduce $\mathbf{J}_2(2\mathbf{q})$ to the form

$$\mathbf{J}_2(2\mathbf{q})=-2i\omega\mathbf{P}_2(2\mathbf{q})=\frac{-\mathbf{q}}{\epsilon(2q,2\omega)}\frac{e^3\hbar^2\omega}{m^2c^2q^2}\sum_{\mathbf{k}}\mathbf{a}_1(\mathrm{SCF})\cdot(\mathbf{k}+\tfrac{1}{2}\mathbf{q})\mathbf{a}_1(\mathrm{SCF})\cdot(\mathbf{k}+\tfrac{3}{2}\mathbf{q})N(\mathbf{k},\mathbf{q},\mathbf{q},\omega,\omega)$$
$$+\frac{\mathbf{q}}{\epsilon(2q,2\omega)}\frac{e^3\omega}{2mc^2q^2}\mathbf{a}_1(\mathrm{SCF})\cdot\mathbf{a}_1(\mathrm{SCF})M(2\mathbf{q},2\omega)\,, \quad (4.6)$$

for $\mathbf{a}_1$(SCF) either longitudinal or transverse.

The induced polarization $\mathbf{P}_2(2\mathbf{q})$ in Eq. (4.6) is the total longitudinal bilinear polarization at frequency 2ω. However, in the SCF calculation the total longitudinal polarization at 2ω is written as

$$\mathbf{q}\cdot\mathbf{P}_2^{\mathrm{SCF}}=\mathbf{q}\cdot\mathbf{P}_2^{\mathrm{SCF}}(\mathrm{Linear})+\mathbf{q}\cdot\mathbf{P}_2^{\mathrm{SCF}}(\mathrm{Nonlinear}), \quad (4.7)$$

where

$$4\pi\mathbf{q}\cdot\mathbf{P}_2^{\mathrm{SCF}}{}_{(\mathrm{Lin.})}=\{\epsilon(2\mathbf{q},2\omega)-1\}(2i\omega/c)\mathbf{q}\cdot\mathbf{a}_2(\mathrm{SCF}). \quad (4.8)$$

Further,

$$(2i\omega/c)\mathbf{q}\cdot\mathbf{a}_2(\mathrm{SCF})+4\pi\mathbf{q}\cdot\mathbf{P}_2^{\mathrm{SCF}}{}_{(\mathrm{Lin})}+4\mathbf{q}\cdot\mathbf{P}_2^{\mathrm{SCF}}{}_{(\mathrm{NL})}=0, \quad (4.9)$$

since there is no external potential at frequency 2ω. Equations (4.7) and (4.9) then lead to

$$\mathbf{J}_2^{\mathrm{SCF}}(2\mathbf{q})=-2i\omega\mathbf{P}_2^{\mathrm{SCF}}(2\mathbf{q})=\frac{\mathbf{J}_2^{\mathrm{SCF}}(\mathrm{NL})}{\epsilon(2\mathbf{q},2\omega)}, \quad (4.10)$$

where $\mathbf{J}_2^{\mathrm{SCF}}(\mathbf{q})$ is longitudinal. General expressions for $\mathbf{J}_2^{\mathrm{SCF}}(\mathrm{NL})$ have been given both in Refs. 1 and 2. For comparison, if we calculate this for our case, we find that $\mathbf{J}_2^{\mathrm{SCF}}(2\mathbf{q})$ is identical with the expression given by Eq. (4.6) which has been obtained in the many-body perturbation theory.

V. DISCUSSION

From the preceding calculations for both the linear and bilinear induced current densities in an electron gas, it is clear that the many-body perturbation theory gives results which are identical with the SCF calculation. However, this has been derived in the pair approximation for the Coulomb interaction. This approximation is valid only for densities which are much higher than electron densities in the most favorable metals. Moreover, in order to treat the case of real metals the uniform positive charge must be replaced by a lattice of positive ions.

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Nuclear Magnetic Resonance in Zintl Intermediate Phases LiCd and LiZn

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The nuclear magnetic resonances have been observed for Li^7 and Cd^{113} in LiCd and for Li^7 and Zn^{67} in LiZn. The Knight shift for $LiCd^{113}$ is 0.39%, for $LiZn^{67}$ 0.20%, for Li^7Cd or Li^7Zn less than 0.01%. An energy-band scheme is proposed for NaTl as well as LiCd and LiZn.

I. INTRODUCTION

THE Zintl phase[1] (*B*-32 structure, Fig. 1) is an ordered intermediate phase ("compound") whose prototype is NaTl. Several other compounds of elements of groups I and III of the periodic table, as well as LiCd and LiZn which are from groups I and II, crystallize in the *B*-32 structure. In this structure, based on the body-centered-cubic (bcc) lattice, the two components each form a diamond lattice, which are interpenetrating. Thus, in the stoichiometric proportion, each atom has eight nearest neighbors, four like and four unlike. It has been proposed[1,2] that in the I–III compounds the alkali metal transfers its valence electron to the heavy metal which then behaves like Ge or Si, forming a diamond lattice. In the I–II compounds, it was proposed[1,2] that an electron is excited from the *d* shell to complete the sp^3 bonding for the heavy metal in the diamond lattice. However, susceptibility measurements[3,4] did not show any signs of the paramagnetism which might be expected to be associated with the *d*-shell excitation. We have observed the nuclear magnetic resonances (NMR) for Li^7 and Cd^{113} in LiCd and for Li^7 and Zn^{67} in LiZn. Again, there is no evidence for *d*-electron excitation. Comparing our results with earlier measurements on[5–8] NaTl and[6] LiAl, LiGa,

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