Influence of exchange effects on the plasmon excitation spectrum of metals: Application in the case of beryllium

N. M. Glezos
Institute of Microelectronics, NCSR "Democritos" P.O. Box 60228, 153.10 Aghia Paraskevi, Attiki, Greece
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The equation-of-motion technique is used to obtain an integral equation that describes collective effects in the homogeneous electron gas, or jellium model of electrons in a solid. This theory should provide the excitation spectrum independently of any external agent and includes exchange effects in the generalized random-phase approximation (GRPA). Collective effects are classified according to their symmetry. Because of the homogeneity one more quantum number is needed in addition to the wave number k, namely, the helicity label m. The plasmon spectrum ($m=0$) is obtained by numerical analysis. Applied to Be, the theory provides a satisfactory interpretation of data obtained by inelastic x-ray scattering. The lower branch appearing in the spectrum of Be is explained in terms of electron short-range interactions from first principles, without the introduction of phenomenological parameters. For those metals for which jellium is a good approximation, it is not necessary to include band effects for the explanation of this second branch.

I. INTRODUCTION

It has been experimentally proved by inelastic x-ray scattering that certain light metals, such as Be,[1-4] Al,[5] Li,[6] and graphite[7] exhibit a double peak in the electron dynamic structure factor $S(\mathbf{q},\omega)$. This peak appears for values of $\mathbf{q}$ inside the particle-hole continuum and for q greater than a critical value.

There are two main reasons for this kind of spectrum, namely, the band structure influence on plasmon states and exchange and correlation effects. Although there is no doubt that both effects influence plasmons, a theory dealing with both at the same time has not been possible yet.

Pandey and co-workers[1,8] as well as Sturm[9] have included band-structure effects in the dielectric response $\epsilon(\mathbf{q},\omega)$ treating a two-band model in the so-called "random-phase approximation including band structure" (RPAB). They have given results for Be (Ref. 1) for a relatively limited region of q, which show a double band for $q\rightarrow 0$ close to the plasmon value.

Brosens, Devreese, and Lemmens[10] calculated the dielectric response function $\epsilon(\mathbf{q},\omega)$ including exchange effects. Green, Nielson, and Szymanski[11] obtained similar results summing up appropriate classes of diagrams in a perturbation expansion giving an explanation of the double peak in the dynamic structure factor of Li and Be.

Awa, Yasuhara, and Asahi[12] calculated the dielectric polarizability including local-field corrections in the Hartree field, thus obtaining a double branch for Al. The second branch is related by them to the dumping effect of one-electron states originating from virtual plasmon emission under the influence of short-range correlations. In their treatment the lower plasmon branch is calculated taking into consideration excitations involving electron-hole pairs and plasmon interactions. Furthermore, they make use of the Hubbard[13] approximation which introduces artificial screening in the electron gas.

Schülke performed experiments using high-resolution synchrotron radiation[14] on monocrystalline Be,[15] and provided arguments in favor of a lattice-induced origin of the double peak based on fitting data to a simple two-band model.

Hong and Lee[16] calculated the dynamic form factor using a method of recurrence relations developed by them[17,18] which involves orthogonal expansion of the density operator. They result in a double-peak structure in the case of Al. Their method has the advantage that the compressibility sum rule and the third-order sum rule are simultaneously satisfied.

Iwamoto, Krotchek, and Pines[19] also calculated the dynamic structure factor introducing pseudopotentials to describe electron interactions, and they attribute the double-peak structure of Be to multipair excitations.

Mukhopadhyay[20] produced similar results by performing a proper linearization of the equation of motion for Wigner distributions demanding a self-consistent calculation which involves the structure factor indirectly.

Zdetsis *et al.*[21,22] attribute the double peak of Be to the splitting of the $3ps$ hybridized band in the band structure of this element calculated by taking clusters of atoms.

In this paper the equation-of-motion technique is used to produce an integral equation which describes the excitation spectrum of the electron gas. Exchange effects are introduced by a proper linearization of the interaction terms known as the generalized random-phase approximation (GRPA).[23-25] The jellium symmetry is taken into consideration, and a more general formalism is developed which predicts nonzero helicity states.[26] However, only the plasmon case is considered, and an application of the theory is made in the case of Be for which extensive experimental data exist. Compared to previous methods both branches of the plasmon spectrum are produced from the same equation and no screening parameters are introduced. Furthermore, the form of the amplitude for each branch gives additional information for its nature.

In Sec. II the formalism is presented and the symmetry properties are taken into account. Various collective states are classified according to the irreducible representations of the full space group. The integral equation which is obtained is examined analytically and numerically in Secs. III and IV, and results are applied to Be. Finally, Sec. V contains concluding remarks.

II. CLASSIFICATION OF COLLECTIVE STATES

We consider the Hamiltonian of a homogeneous N-electron system subject to an external potential $U(\mathbf{r})$ expressed in the second quantization representation:

$$\hat{\mathcal{H}}=\tfrac{1}{2}\int \nabla\Psi^{\dagger}(\mathbf{r})\nabla\Psi(\mathbf{r})d\mathbf{r} + \tfrac{1}{2}\int \frac{\Psi^{\dagger}(\mathbf{r})\Psi^{\dagger}(\mathbf{r}')\Psi(\mathbf{r}')\Psi(\mathbf{r})}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}\,d\mathbf{r}' + \int U(\mathbf{r})\Psi^{\dagger}(\mathbf{r})\Psi(\mathbf{r})d\mathbf{r} \ . \tag{2.1}$$

In this equation the spin index has been suppressed since we are interested in the spectrum of spin-zero states. The formalism presented here is used to deduce the properties of the spin-zero states of the electron gas. In Eq. (2.1) the Atomic Unit System is used. By $\Psi^{\dagger}(\mathbf{r}),\Psi(\mathbf{r})$ we mean the usual creation and annihilation fields which obey the following anticommutation relations:

$$\{\Psi^{\dagger}(\mathbf{r}),\Psi^{\dagger}(\mathbf{r}')\}_{+}=0 \ , \tag{2.2a}$$

$$\{\Psi(\mathbf{r}),\Psi(\mathbf{r}')\}_{+}=0 \ , \tag{2.2b}$$

$$\{\Psi^{\dagger}(\mathbf{r}),\Psi(\mathbf{r}')\}_{+}=\delta(\mathbf{r}-\mathbf{r}') \ . \tag{2.2c}$$

The symmetry group of (2.1) has already been discussed.[26] It has been proved that with $U(\mathbf{r})=0$ the Hamiltonian (2.1) is invariant under the full space group I_3^3. This group consists of all translations and rotations in the three-dimensional Euclidean space, and it is a six-parameter Lie group. The eigenstates of (2.1) in the absence of an external potential $U(\mathbf{r})$ can be classified according to the irreducible representations of the group. These are labeled by the eigenvalues of its two Casimir operators $\mathbf{P}^2$ and $\mathbf{L}\cdot\mathbf{P}$, where $\mathbf{P}$ denotes the total momentum and $\mathbf{L}$ the total angular momentum of the N-particle system. By $\mathbf{L}\cdot\mathbf{P}$ we denote the helicity operator. In the second quantization representation operators $\mathbf{P}$ and $\mathbf{L}$ have the form

$$\mathbf{P}=\int \mathbf{j}(\mathbf{r})d\mathbf{r} \ , \tag{2.3}$$

$$\mathbf{L}=\int \mathbf{r}\times\mathbf{j}(\mathbf{r})d\mathbf{r} \ , \tag{2.4}$$

where $\mathbf{j}(\mathbf{r})$ is the current operator,

$$\mathbf{j}(\mathbf{r})=\frac{1}{2i}[\Psi^{\dagger}(\mathbf{r})\nabla\Psi(\mathbf{r})-\nabla\Psi^{\dagger}(\mathbf{r})\Psi(\mathbf{r})] \ . \tag{2.5}$$

In order to classify the eigenvalues, one must form realizations of this group in an appropriate function space. This has already been carried out for various spaces.[27] The most appropriate realization for our physical problem is that of the two-particle function space. For example, the basis

$$\Phi^{q,m}_{k_z,k_\perp}(\mathbf{r},\mathbf{R})=(-i)^m J_m(k_\perp r_\perp) \times\exp[i(k_z+q/2)r_z+im\phi_r+i\mathbf{q}\cdot\mathbf{R}] \tag{2.6}$$

transforms according to the $\mathbf{q},m$ irreducible representation of the group I_3^3. Here $\mathbf{r},\mathbf{R}$ are the center-of-mass coordinates for two identical particles $\mathbf{r}=\mathbf{r}_1-\mathbf{r}_2$ and $\mathbf{R}=(\mathbf{r}_1+\mathbf{r}_2)/2$. By J_m we denote the ordinary cylindrical Bessel function of order m and $\mathbf{k},\mathbf{r}$ are expressed in cylindrical coordinates $\mathbf{k}=(k_z,k_\perp,\phi_{\mathbf{k}})$ and $\mathbf{r}=(r_z,r_\perp,\phi_{\mathbf{r}})$. The vector $\mathbf{q}$ defines the z axis of the coordinate system, and so the various directions of $\mathbf{q}$ define the elements of the irreducible subspace. The meaning of labels q and m is clarified by application of the operators $\mathbf{P}^2$, $\mathbf{L}\cdot\mathbf{P}$, and $\mathbf{P}$ on (2.6):

$$\mathbf{P}^2\Phi^{q,m}_{k_z,k_\perp}=q^2\Phi^{q,m}_{k_z,k_\perp} \ , \tag{2.7}$$

$$\mathbf{L}\cdot\mathbf{P}\Phi^{q,m}_{k_z,k_\perp}=mq\Phi^{q,m}_{k_z,k_\perp} \ , \tag{2.8}$$

$$\mathbf{P}\Phi^{q,m}_{k_z,k_\perp}=\mathbf{q}\Phi^{q,m}_{k_z,k_\perp} \ . \tag{2.9}$$

From the previous relations it is evident that $\mathbf{q}$ is the momentum eigenvalue and m the helicity eigenvalue, i.e., the projection of angular momentum along $\mathbf{q}$.

Despite its apparent simplicity, basis (2.6) has the defect that the coordinate system is defined by $\mathbf{q}$ along the z axis. Even so it can be used for the expansion of two-particle field operators as follows:

$$\Psi^{\dagger}(\mathbf{R}+\mathbf{r}/2)\Psi(\mathbf{R}-\mathbf{r}/2)=\sum_{k_z,k_\perp} A^{q,m}_{k_z,k_\perp}\cdot\Phi^{q,m}_{k_z,k_\perp}(\mathbf{r},\mathbf{R}) \ . \tag{2.10}$$

Operators $A^{q,m}_{k_z,k_\perp}$ defined by (2.10) have the symmetry described above. They will be used to generate the eigenstates of the Hamiltonian. Operators such as Fourier transforms of the density and current operators are expressed in a simple manner. Using $\hat{\mathbf{q}}$ to denote a unit vector along $\mathbf{q}$ and

$$\hat{\mathbf{q}}_{\pm}=(\hat{\mathbf{q}}_x\pm i\hat{\mathbf{q}}_y)/\sqrt{2} \ , \tag{2.11}$$

we find,

$$\rho^{\dagger}_{\mathbf{q}}=\sum_{k_\perp,k_z} A^{q,0^{\dagger}}_{k_\perp p,k_z} \ , \tag{2.12}$$

$$\hat{\mathbf{q}}\cdot\mathbf{J}_{\mathbf{q}}=\sum_{k_\perp,k_z}(k_z+q/2)A^{q,0^{\dagger}}_{k_\perp,k_z} \ , \tag{2.13}$$

$$\hat{\mathbf{q}}_{\pm}\cdot\mathbf{J}_{\mathbf{q}}=\sum_{k_\perp,k_z}\frac{k_\perp}{\sqrt{2}}A^{q,\pm 1^{\dagger}}_{k_\perp,k_z} \ . \tag{2.14}$$

So (2.11) denotes the right- and left-circularly polarized components of current, a representation appropriate for the description of transverse and circularly polarized states. In order to obtain the excitation spectrum, we construct the equation of motion for the two-particle operator in (2.10):

$$[\hat{\mathcal{H}}, A^{\mathbf{q},m}_{k_\perp,k_z}] = \left[\frac{\mathbf{q}^2}{2}+\mathbf{k}\cdot\mathbf{q}\right] A^{\mathbf{q},m}_{k_\perp,k_z} + \sum_{\mathbf{k}'}\frac{2\pi}{\mathbf{k}'^2}\frac{1}{2\pi}\int_0^{2\pi}\exp(im\phi^{\mathbf{q}}_{\mathbf{k}})[(a^\dagger_{\mathbf{k}-\mathbf{k}'+\mathbf{q}}a_{\mathbf{k}}-a^\dagger_{\mathbf{k}+\mathbf{q}}a_{\mathbf{k}+\mathbf{k}'})\rho^\dagger_{\mathbf{k}'}+\rho^\dagger_{\mathbf{k}'}(a^\dagger_{\mathbf{k}-\mathbf{k}'+\mathbf{q}}a_{\mathbf{k}}-a^\dagger_{\mathbf{k}+\mathbf{q}}a_{\mathbf{k}+\mathbf{k}'})]d\phi^{\mathbf{q}}_{\mathbf{k}} . \quad (2.15)$$

Expression (2.15) is linearized by performing a Hartree-Fock linearization known as GRPA. In this approximation pairings of creation and annihilation operators are formed and only those corresponding to direct and exchange scattering of the electrons are kept.[23–26] The final result is

$$[\hat{\mathcal{H}}, A^{\mathbf{q},m}_{k_z,k_\perp}] = [E_{\mathrm{HF}}(\mathbf{k}+\mathbf{q})-E_{\mathrm{HF}}(\mathbf{k})]A^{\mathbf{q},m}_{k_z,k_\perp} + \delta_{m,0}(n_{\mathbf{k}}-n_{\mathbf{k}+\mathbf{q}})\frac{4\pi}{q^2}\sum_{k_z',k_\perp'}A^{\mathbf{q},m}_{k_z',k_\perp'} - 4\pi(n_{\mathbf{k}}-n_{\mathbf{k}+\mathbf{q}})\sum_{k_z',k_\perp'}\left[\frac{1}{2\pi}\int_0^{2\pi}\frac{\exp(im\phi^{\mathbf{q}}_{k})}{|\mathbf{k}-\mathbf{k}'|^2}d\phi^{\mathbf{q}}_{\mathbf{k}}\right]A^{\mathbf{q},m}_{k_z',k_\perp'} , \quad (2.16)$$

where

$$E_{\mathrm{HF}}(k)=\frac{k^2}{2}+\sum_{\mathbf{k}'}\frac{4\pi n_{k'}}{|\mathbf{k}-\mathbf{k}'|^2} \quad (2.17)$$

is the Hartree-Fock energy contributing as a self-energy term and $n_{\mathbf{k}}$ is the zero-temperature ground-state occupation function for the free-electron system:

$$n_{\mathbf{k}}=\begin{cases}1, & k\le k_F\\ 0, & k>k_F\end{cases} . \quad (2.18)$$

Excited states having a $\mathbf{q},\mathbf{m}$ symmetry are produced by applying an operator $B^m_{\mathbf{q}}$ on the ground state $|\Psi_0\rangle$. If the commutator of $B^m_{\mathbf{q}}$ is of the form

$$[\hat{\mathcal{H}},B^m_{\mathbf{q}}]=\omega^m_{\mathbf{q}}B^m_{\mathbf{q}} , \quad (2.19)$$

then $\omega^m_{\mathbf{q}}$ is the excitation energy from state $|\Psi_0\rangle$ state $B^m_{\mathbf{q}}|\Psi_0\rangle$.

Examing the linearized expression (2.16), we see that $B^m_{\mathbf{q}}$ can be selected in the form of a linear product of $A^{\mathbf{q},m}_{k_z,k_\perp}$:

$$B^m_{\mathbf{q}}=\sum_{k_z,k_\perp}f^m_{\mathbf{q}}(k_z,k_\perp)A^{\mathbf{q},m}_{k_z,k_\perp} . \quad (2.20)$$

Using (2.20) in connection with (2.16), we can see that the commutation relation (2.19) is approximately satisfied in the frame of the GRPA if and only if the amplitude $f^m_{\mathbf{q}}(k_z,k_\perp)$ obeys the following integral equation:

$$[\omega^m_{\mathbf{q}}-E_{\mathrm{HF}}(\mathbf{k}+\mathbf{q})+E_{\mathrm{HF}}(\mathbf{k})]f^m_{\mathbf{q}}(k_z,k_\perp) = \delta_{m,0}\frac{1}{\pi^2q^2}\int(n_{\mathbf{k}'}-n_{\mathbf{k}'+\mathbf{q}})f^m_{\mathbf{q}}(k_z',k_\perp')d\mathbf{k}'+\frac{1}{\pi^2}\int(n_{\mathbf{k}'+\mathbf{q}}-n_{\mathbf{k}'})\frac{\exp[im(\phi^{\mathbf{q}}_{\mathbf{k}}-\phi^{\mathbf{q}}_{\mathbf{k}'})]}{|\mathbf{k}-\mathbf{k}'|^2}f^m_{\mathbf{q}}(k_z,k_\perp)d\mathbf{k}' . \quad (2.21)$$

Solutions of (2.21) for $m\neq 0$ have been discussed. Localized solutions are particle-hole pairs of momentum $\mathbf{q}$ accompanied by a polarization cloud of well-defined helicity m.

Extended solutions also exist. They are low-lying energy states of momentum $\mathbf{q}$ and helicity m similar to the zero-sound-mode states obtained in the frame of Fermi-liquid theory. These states are Landau damped. However (to our knowledge), no attempt has been made for the experimental confirmation of the existence of such states.

The $m=0$ solutions of (2.21) appear in electron-energy-loss and x-ray spectra. These solutions are discussed in connection with appropriate experiments and existing theories.

First, we rewrite Eq. (2.21) for $m=0$ in a dimensionless form:

$$[\Omega-\Delta E(Q;x,\mu)]F(Q;x,\mu)=\frac{4\beta}{Q^2}\int_0^{+\infty}x'^2dx'\int_{-1}^{1}d\mu'(n_{\mathbf{x}'}-n_{\mathbf{x}'+\mathbf{Q}})F(Q;x',\mu') - 4\beta\int_0^{+\infty}x'^2dx'\int_{-1}^{1}d\mu'(n_{\mathbf{x}'}-n_{\mathbf{x}'+\mathbf{Q}})\left[\frac{1}{2\pi}\int_0^{2\pi}\frac{d\phi_{\mathbf{x}'}}{|\mathbf{x}-\mathbf{x}'|^2}\right]F(Q;x',\mu') , \quad (2.22)$$

where $\Omega=\omega_q^0/\varepsilon_F$, $\mathbf{x}=\mathbf{k}/k_F$, $\mathbf{Q}=\mathbf{q}/k_F$, and $\mu=\cos\vartheta_k$ with ϵ_F the Fermi energy and k_F the Fermi wavelength. Although the symmetry arguments suggest cylindrical coordinates for $\mathbf{k}$, x,μ are more appropriate variables in Eq. (2.22).

The solution $F(Q;x,\mu)$ is expressed in terms of these coordinates and a dimensionless parameter β, which is a measure of the ratio between potential and kinetic energy in units of the Wigner-Seitz radius r_s of the electron gas:

$$\beta=\frac{e^2k_F}{2\pi\epsilon_F}=0.1658r_s \ . \tag{2.23a}$$

Then

$$\Delta E(Q;x,\mu)=Q^2+2Qx\mu-\beta g(|\mathbf{x}+\mathbf{Q}|)+\beta g(x) \ , \tag{2.23b}$$

with

$$g(x)=\frac{1-x^2}{x}\ln\left|\frac{x+1}{x-1}\right|+2 \ . \tag{2.23c}$$

In terms of β the plasmon energy reads

$$\Omega_p=\omega_p/\epsilon_F=\sqrt{16\beta/3} \ . \tag{2.23d}$$

Brosens and Devreese,[28] dealing with the dynamical exchange effects in the dielectric function, produced an equation for the amplitude similar to (2.22). They developed a numerical procedure for its solution based on vectorization of the integral equation, and they derived results which included exchange corrections in the spectrum of the particle-hole continuum. Although these authors mention the possibility of an iterative approach, they do not use it because of the expected oscillatory behavior of the solutions. This is due to the fact that the integral equation provides solutions both for particle-hole excitations and collective excitations.

Hong and Lee[16] calculated the dynamic form factor by developing a method based on recurrence relations.[17,18] In their approach they have proved that the GRPA leading to (2.22) is not satisfactory since it cannot obey simultaneously both the compressibility and the third-order frequency-moment sum rules. In calculating the dynamic form factor, no double peak appears if one does not go further than the GRPA.[17,19]

The method used in this paper differs from the dynamical RPA approach in that a direct search is made for the energy spectrum of the collective oscillations and not through the dynamical form factor. This is achieved by using an iterative procedure with the long-wavelength collective solutions as a starting point and q as an iterative parameter. Then solutions for finite q are obtained by extending the trial solutions with the assumption of continuity with respect to q. Details are presented in the next section.

III. LONG-WAVELENGTH STATES

With a complicated integral equation like (2.22), it is useful to begin with a limiting case that can be handled by analytic methods. The results form a starting point for a full numerical solution.

In the long-wavelength limit (2.22) simplifies because states which participate in collective effects are concentrated in a region close to the Fermi surface. Thus, when the reduced wave vector $Q\to 0$, we have

$$n_{\mathbf{x}}-n_{\mathbf{x}+\mathbf{Q}}\to\mu Q\delta(x-1) \ , \tag{3.1}$$

and

$$\Omega-\Delta E(Q;x,\mu)\to\Omega-2Q\mu \ , \tag{3.2}$$

when the self-energy terms are neglected in the energy difference ΔE. The kernel of the integral equation is different from zero only on the Fermi surface. We expand the solution $F(Q;x,\mu)$ for $x=1$, in Legendre polynomials of the first kind,

$$F(Q;1,\mu)=\sum_n c_nP_n(\mu) \ , \tag{3.3}$$

and the Coulomb kernel in a double series of spherical harmonics. The problem of the vanishing denominator in (2.22) is treated by taking the limit $x\to 1\pm$ in both sides of the integral equation. These limits coincide. After neglecting the self-energy terms, we finally obtain the following linear system for the coefficients c_n:

$$c_n=c_1\frac{2n+1}{3}\frac{4\beta}{Q^2}G^n_{000}(\zeta)-2\beta(2n+1)\sum_{n'}c_{n'}\sum_{LL'M}\lambda^{n'}_{LL'M}G^{n'}_{LL'M}(\zeta) \ , \tag{3.4}$$

with

$$G^n_{LL'M}(\zeta)=\int\frac{P_n(\mu)Y_{LM}(\hat{\mathbf{x}})Y^*_{L'M}(\hat{\mathbf{x}})}{\zeta-\mu}d\hat{\mathbf{x}} \ , \tag{3.5a}$$

$$\zeta=\frac{\Omega}{2Q}=\frac{\omega}{qk_F} \ , \tag{3.5b}$$

and

$$\lambda^n_{LL'M}=\frac{\sqrt{\pi}}{2(2n+1)(2L+1)(2L'+1)}\left[\frac{n+1}{\sqrt{2n+3}}D^{n+1}_{LL'M}+\frac{n}{\sqrt{2n+1}}D^{n-1}_{LL'M}\right] \ , \tag{3.5c}$$

where D denotes the usual Clebsch-Gordan coefficients. Equation (3.4) can be solved numerically.

An alternative procedure is to attempt solution by iteration since in the long-wavelength limit the Hartree potential dominates over the exchange interactions. Thus, as a starting point for iteration, we select the plasmon solution in the absence of exchange. Neglecting the second term in the right term of (3.4) yields the plasmon solution

$$c_n^{(0)}=c_1^{(0)}\frac{4\beta}{Q^2}\mathcal{Q}_n(\zeta)\ , \tag{3.6}$$

since $G^n_{000}(\zeta)$ is the Legendre function of the second kind $\mathcal{Q}_n(\zeta)$. Substituting (3.6) in the expansion (3.3) gives the known solution, namely,

$$F^{(0)}(Q;1,\mu)=\frac{a(Q)}{\Omega-2Q\mu}\ , \tag{3.7}$$

where $a(Q)$ is an arbitrary function of Q.

In order to examine the influence of the exchange terms on the plasmon spectrum, (3.6) is substituted in (3.4) to yield the next step of iteration. To find an approximate dispersion relation, one equates coefficients in the right and the left members and keeps the less oscillating part of the exchange term. This gives

$$1-\frac{4\beta}{Q^2}\mathcal{Q}_1(\zeta)+\beta\left[\frac{1}{\sqrt{3}}\mathcal{Q}_1(\zeta)+\frac{1}{5}\mathcal{Q}_2(\zeta)\right]\simeq 0\ . \tag{3.8}$$

For plasmon states in the limit $Q\to 0$ or equivalently $\zeta\to+\infty$ with Ω nonzero, the Legendre functions $\mathcal{Q}_1,\mathcal{Q}_2$ can be approximated as power series in $1/\zeta$. The final result for the plasmon dispersion is

$$\Omega=\Omega_p+\frac{6Q^2}{5\Omega}(1-0.068r_s)\ . \tag{3.9}$$

This result is similar to that derived by Kanazawa and Tani[29] using perturbation expansion methods and by other authors.[10,29–39] They obtain a correction factor $\Delta=1-0.055r_s$, which gives for Be a coefficient 0.42 for the Q^2 term, while (3.9) gives 0.41, in coincidence with the experimental value.

The procedure described serves as a starting point for obtaining solutions for finite values of Q. The necessary steps are as follows.

(i) For small Q we solve (3.4) numerically or by iteration and obtain the solution on the Fermi surface $F(Q;1,\mu)$.

(ii) This is substituted in the second member of (2.22) taking also the limiting value given by (3.1) to yield $F(Q;x,\mu)$.

(iii) Integration of (2.22) over phase space x,μ produces a dispersion relation which is solved numerically to obtain the values of Ω which permit a solution of (2.22) for given Ω.

(iv) Starting from these values for Ω and $F(Q;x,\mu)$ and augmenting Q by a small step $Q'=Q+\Delta Q$, one can substitute in (2.22) to find the corresponding solution Ω' and $F(Q';x,\mu)$.

IV. INTERMEDIATE WAVE VECTORS: RESULTS FOR Be

A brute force attempt to solve (2.22) might result in solutions not having direct physical significance. An acceptable solution can be obtained by demanding consistency of numerical solutions with those obtained analytically in the long-wavelength limit.

In order to obtain the plasmon-type spectrum for finite values of Q, we start from the long-wavelength limit and extend the solutions in the particle-hole continuum. The first step is to obtain an appropriate dispersion relation for the spectrum. This is achieved by multiplying (2.22) with $n_{\mathbf{x}}-n_{\mathbf{x}+\mathbf{Q}}$ and integrating over phase space. After a few manipulations the following dispersion relation is obtained:

$$A(Q;\Omega)=A_0(Q;\Omega)+A_{\rm exc}(Q;\Omega)=0\ , \tag{4.1}$$

where

$$A_0(Q;\Omega)=1-\frac{4\beta}{Q^2}\int_0^{+\infty}x^2dx\int_{-1}^{+1}d\mu\frac{n_{\mathbf{x}}-n_{\mathbf{x}+\mathbf{Q}}}{\Omega-\Delta E(Q;x,\mu)}\ , \tag{4.2}$$

and

$$A_{\rm exc}(Q;\Omega)=4\beta\int_0^{+\infty}x^2dx\int_0^{+\infty}x'^2dx'\int_{-1}^{+1}d\mu\int_{-1}^{+1}d\mu' \times\frac{(n_{\mathbf{x}}-n_{\mathbf{x}+\mathbf{Q}})(n_{\mathbf{x}'}-n_{\mathbf{x}'+\mathbf{Q}})}{\Omega-\Delta E(Q;x,\mu)}\left[\frac{1}{2\pi}\int_0^{2\pi}\frac{d\phi_{\mathbf{x}'}}{|\mathbf{x}'-\mathbf{x}|^2}\right]F(Q;x',\mu')\ . \tag{4.3}$$

In order to simplify (4.1), $F(Q;x,\mu)$ has been normalized posing the condition

$$\int_0^{+\infty}x^2dx\int_{-1}^{+1}d\mu(n_{\mathbf{x}}-n_{\mathbf{x}+\mathbf{Q}})F(Q;x,\mu)=1\ . \tag{4.4}$$

The $A_0(Q;\Omega)$ contribution to the dispersion relation in the long-wavelength limit takes the form (3.8), which has already been discussed. It is similar in form to the known RPA dispersion relation with additional exchange terms in the denominator. The $A_{\text{exc}}(Q;\Omega)$ part of the dispersion relation contains short-range interactions, and its importance for various values of r_s is discussed below.

In Figs. 1–3 both $A_0(Q;\Omega)$ and $A(Q;\Omega)$ are plotted in the case of Be ($r_s=1.88$) for different values of q as a function of the reduced frequency variable $\zeta=\Omega/2Q$. For small values of q, e.g., for $q=0.2k_F$ in Fig. 1, the roots of $A_0(Q;\Omega)$ and $A(Q;\Omega)$ are close. Only one of the two possible roots appears in the figure. The root value of ζ, which gives the plasmon spectrum whose limiting expression is given by (3.4), is too large to be included in the figure. When q approaches the critical value $q_c=0.78k_F$ in Fig. 2, the function $A_0(Q;\Omega)$ touches the ζ axis and we have a single root. For greater values of q, $A_0(Q;\Omega)$ has no real root. This is not the case when exchange is included in (4.1). The plotting of $A(Q;\Omega)$ in Figs. 1–3 for the same values of q reveals that we still have two solutions correspondings to two branches in the plasmon spectrum, thereafter denoted by Ω_- and Ω_+ (lower and upper branches). Also, a discontinuity appears in $A(Q;\Omega)$.

In Fig. 4 the two branches of the spectrum has been plotted for the case of Be together with available experimental data. There is good agreement between the present theory and experimental data with the lower branch, but the observed structure around $q\simeq 2.0k_F$ in the upper branch is not reproduced by the theory. This is probably an indication that when the value of q^{-1} approaches interatomic distances the jellium model breaks down and lattice effects become important. All the same, the method presented leads to a qualitatively correct prediction for the plasmon spectrum.

The lower branch Ω_- represents longitudinal collective excitations of a different nature than the plasmons. As it will be seen by examining the amplitude $F(Q;x,\mu)$ for both branches, plasmon states are connected to deformations extended over the entire Fermi surface, while energy states belonging to the lower branch correspond to deformations localized in wave-vector space, and they are consequently Landau damped.

By examining the dispersion relation (4.1), we find that when q is of the order of k_F there is no real solution for $r_s<1.7$. On the other hand, when r_s is greater than 4.3 and for the same region of q values, the model predicts one single branch, that corresponding to Ω_+. In the intermediate region $1.7<r_s<4.3$, both branches are expected. It is to be noted that all three metals Al, Be, and Li, for which the lower branch has been observed experimentally, have r_s in this range. It is interesting to carry

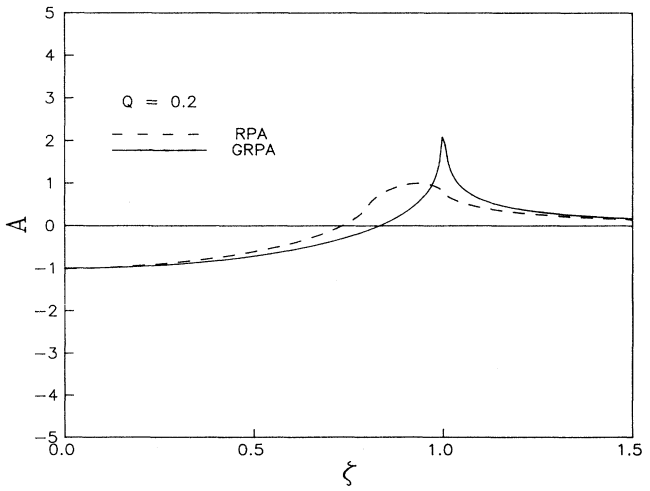

FIG. 1. $A_0(Q;\Omega)$ (dashed line) and $A(Q;\Omega)$ (solid line) vs the reduced variable $\zeta=\Omega/2Q$ in the case of Be ($r_s=1.88$) for $Q=q/k_F=0.2$. Both functions intersect the ζ axis in two points, but only one appears in the figure.

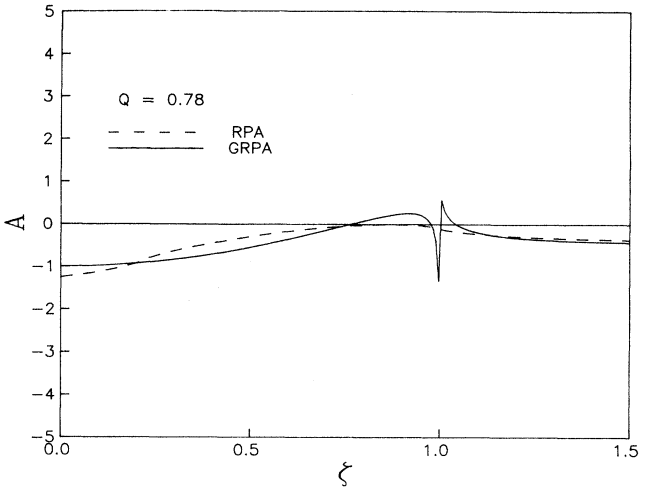

FIG. 2. As in Fig. 1 for $Q=0.78$ close to the critical value. $A_0(Q;\Omega)$ touches the ζ axis, while $A(Q;\Omega)$ has two roots and a discontinuity appears.

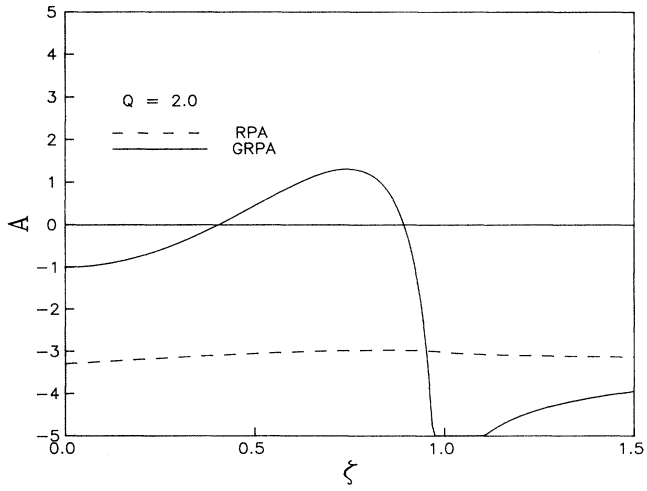

FIG. 3. As in Fig. 1 for $Q=2.0$. $A_0(Q;\Omega)$ has no real roots.

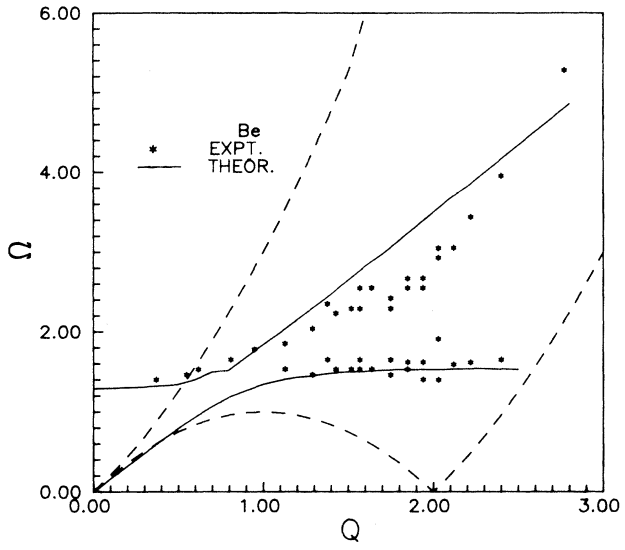

FIG. 4. Excitation energy $\Omega=\omega/E_F$ vs reduced wave vector $Q=q/k_F$. The solid line represents theory, while asterisks stand for the experiment of Vradis and Priftis (Ref. 4). Dashed lines mark the limits of particle-hole spectrum.

out detailed experiments on other metals having r_s in this range and for which the jellium model is a good approximation in order to check the existence of the predicted lower branch.

The different nature of the two branches becomes evident from the shape of the amplitude in Figs. 5–7, which is plotted for the same values of q and their corresponding Ω. Phase space is described by the variables x,μ as defined in connection to (2.22). The plotted function in these figures is $\Phi(Q;x,\mu)$, defined by

$$\Phi(Q;x,\mu)=[\Omega-\Delta E(Q;x,\mu)]F(Q;x,\mu)\ , \tag{4.5}$$

where $F(Q;x,\mu)$ is normalized by condition (4.4). The

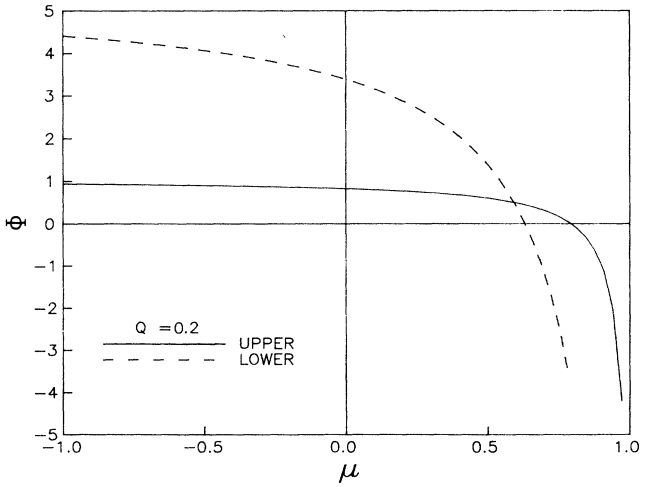

FIG. 5. Amplitudes Φ given by (4.5) for $x=1$ and $Q=0.2$ for the corresponding Ω_- (dashed line) and Ω_+ (solid line) vs $\mu=\cos\vartheta_{\mathbf{k}}$ in the case of Be.

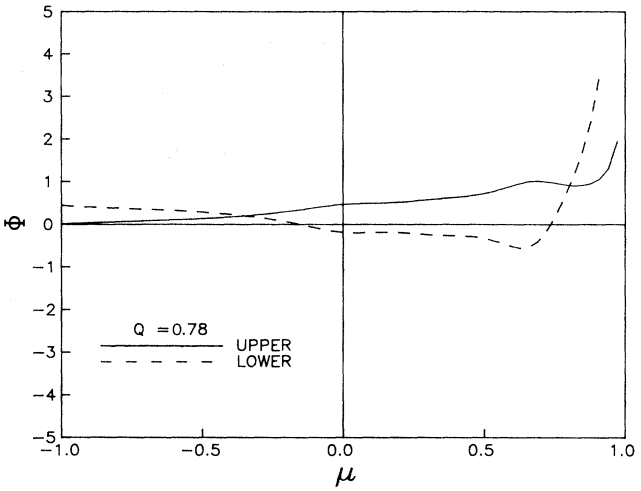

FIG. 6. As in Fig. 5 for $Q=0.78$.

values of $\mathbf{k}$ which lead to the collective plasmon mode Ω_+ appear to be extended over the entire Fermi surface. In particular, for values of q inside the particle-hole continuum, localizations coexist with an overall deformation of the surface. The major contribution to the amplitude comes from $\mathbf{k}$ along $\mathbf{q}$, i.e., in the direction of plasmon propagation $\mu=\cos\vartheta_{\mathbf{k}}=1$. The lower branch Ω_- is of a different nature. The amplitude $\Phi(Q;x,\mu)$ is localized around a certain $\mathbf{q}$-dependent direction of $\mathbf{k}$. Thus the longitudinal wave has a finite lifetime depending on the localization width and decays into particle-hole excitations.

V. CONCLUSIONS

In this paper the dynamical Hartree-Fock equation was derived as the zero helicity case of a more general equation describing the excitation spectrum of the elec-

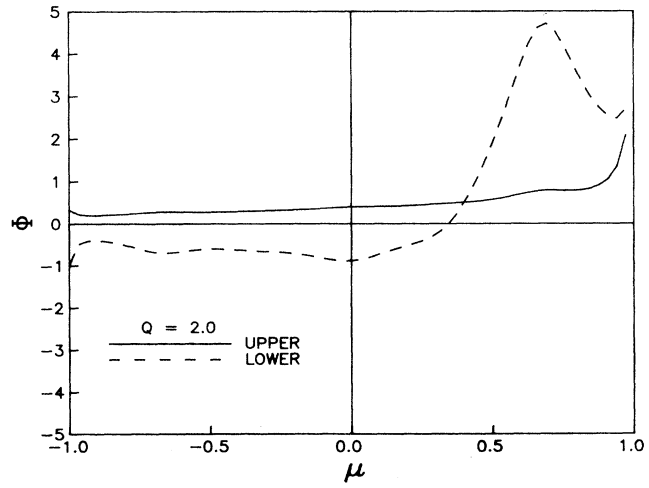

FIG. 7. As in Fig. 5 for $Q=2.0$.

tron gas.

For values of r_s between 1.7 and 4.3, two branches of collective excitations are predicted, one being the plasmon branch. The lower branch has been observed experimentally in certain metals with r_s in the predicted range. Yasuhara, and Asahi[12] have also dealt theoretically with the lower branch by considering excitations involving electron-hole pairs and plasmon interactions.

The upper branch solutions coincide numerically with previous theoretical results in the long-wavelength limit.

Finally, we would like to comment that the origin of the experimentally observed lower branch is due to collective excitations in crystals for which the jellium model is a good approximation. In other cases the crystal structure cannot be neglected.

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[1]K. C. Pandey, P. M. Platzman, P. Eisenberger, and E-Ni Foo, Phys. Rev. B **9**, 5046 (1974).
[2]J. Boviatsis and G. D. Priftis, Solid State Commun. **33**, 577 (1980).
[3]G. D. Priftis and J. Boviatsis, Phys. Status Solidi B **104**, 673 (1981).
[4]A. Vradis and J. D. Priftis, Phys. Rev. B **32**, 3556 (1984).
[5]E. Petri, A. Otto, and W. Hanke, Solid State Commun. **19**, 711 (1976).
[6]G. D. Priftis, J. Boviatsis, and A. Vradis, Phys. Lett. **68A**, 482 (1978).
[7]P. M. Platzman and P. Eisenberger, Phys. Rev. Lett. **31**, 152 (1974).
[8]P. Eisenberger, P. M. Platzman, and K. C. Pandey, Phys. Rev. Lett. **31**, 311 (1973).
[9]K. Sturm, Z. Phys. B **29**, 27 (1978).
[10]F. Brosens, J. T. Devreese, and L. F. Lemmens, Phys. Rev. B **21**, 1363 (1980); **21**, 1349 (1980); Phys. Status Solidi B **74**, 45 (1976).
[11]F. Green, D. Nielson, and J. Szymanski, Phys. Rev. B **31**, 2796 (1985); **31**, 2796 (1985).
[12]K. Awa, H. Yasuhara, and T. Asahi, Solid State Commun. **38**, 1285 (1981); Phys. Rev. B **25**, 3670 (1982); **25**, 3687 (1982).
[13]J. Hubbard, Proc. R. Soc. London Ser. A **243**, 336 (1957).
[14]W. Schülke, Nucl. Instrum. Methods Phys. Res. A **246**, 491 (1986).
[15]W. Schülke, U. Bonse, H. Nagasawa, S. Mourikis, and A. Kaprolat, Phys. Rev. Lett. **59**, 1361 (1987).
[16]J. Hong and M. H. Lee, Phys. Rev. Lett. **55**, 2375 (1985).
[17]J. Hong, J. Park, and M. H. Lee, Phys. Rev. B **40**, 1528 (1989).
[18]M. H. Lee, Phys. Rev. B **26**, 2547 (1982).
[19]N. Iwamoto, E. Krotchek, and D. Pines, Phys. Rev. B **29**, 3924 (1984); **29**, 3936 (1984).
[20]G. Mukhopadhyay, Phys. Scr. **38**, 224 (1988).
[21]A. D. Zdetsis and D. Miliotis, Solid State Commun. **42**, 227 (1982).
[22]Z. D. Zdetsis and D. Papademitriou, Phys. Rev. Lett. **60**, 61 (1988).
[23]D. Pines and Ph. Nozières, *The Theory of Quantum Liquids* (Benjamin, New York, 1966).
[24]P. M. Platzman and P. A. Wolff, *Solid State Physics Supplement 13* (Academic, New York, 1966).
[25]S. Ichimaru, Rev. Mod. Phys. **34**, 1017 (1982).
[26]N. M. Glezos and A. K. Theophilou, Physica A **146**, 360 (1987).
[27]W. Miller, *Lie Theory and Special Functions* (Academic, New York, 1968); Commun. Pure Appl. Math. **42**, 567 (1964); **43**, 493 (1965); **43**, 679 (1985).
[28]F. Brosens and T. Devreese, Phys. Status Solidi B **147**, 173 (1988).
[29]H. Kanazawa and Sho-ichiro Tani, Prog. Theor. Phys. **19**, 153 (1958).
[30]R. Ferrel, Phys. Rev. **107**, 450 (1957).
[31]V. P. Silin, J.E.T.P. **34**, 781 (1958).
[32]H. Kanazawa, S. Misawa, and E. Fujita, Prog. Theor. Phys. **23**, 426 (1960).