Hydrodynamic analysis of coupled plasmons
by David Leo Burdick

A thesis submitted to the Graduate Faculty in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Physics
Montana State University

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A hydrodynamic approach has been combined with the equation of motion method to treat the nonlinear interactions of plasmons in an electron gas. Exchange and correlation effects are included by means of appropriate pressure terms. Corrections to the K2term of the plasmon energy are obtained by solving the integral eigenvalue equation found for the new collective excitation energy. It is found that the new plasmon energy becomes imaginary when the density parameter rs=16.9; this is a possible indication that the electron gas is undergoing a phase transition into the electron lattice. It is found that the changes in the ground state energy caused by the new plasmon energy reduce the ground state energy by about 10% for the alkali metals, worsening the agreement with experiment. The effects of plasmon-plasmon coupling do not appear to be experimentally observable either in transmission experiments or experiments dependent on plasmon lifetimes.

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ABSTRACT

A hydrodynamic approach has been combined with the equation of motion method to treat the nonlinear interactions of plasmons in an electron gas. Exchange and correlation effects are included by means of appropriate pressure terms. Corrections to the k^2 term of the plasmon energy are obtained by solving the integral eigenvalue equation found for the new collective excitation energy. It is found that the new plasmon energy becomes imaginary when the density parameter r_s=16.9; this is a possible indication that the electron gas is undergoing a phase transition into the electron lattice. It is found that the changes in the ground state energy caused by the new plasmon energy reduce the ground state energy by about 10% for the alkali metals, worsening the agreement with experiment. The effects of plasmon-plasmon coupling do not appear to be experimentally observable either in transmission experiments or experiments dependent on plasmon lifetimes.

I. INTRODUCTION

That branch of physics commonly called many-body theory is now about 20 years old. Many-body theory deals with the problem of describing systems which contain many interacting particles (perhaps 10^{23} particles per cubic centimeter is the order of magnitude).

The classical problem in modern many-body theory is that of interacting electrons in a metal. The long-range nature of the Coulomb interaction between electrons prevented a consistent description of electrons in metals for many years. The usual Rayleigh-Schrodinger perturbation techniques are not powerful enough to cope with the divergences which appear when one tries to treat the Coulomb interaction as a small perturbation on the kinetic energy.

The first truly successful attempt to incorporate the Coulomb interaction into the Hamiltonian for an electron system in a metal was carried out by Bohm and Pines in a series of papers in the early fifties.[1] The essential feature of their treatment (which is usually called the collective-coordinates description) was the separation of the long range and short-range interactions; the long-range part gives rise to the collective oscillations in the system which are called plasmons. The short-range portion describes

the motion of the individual electrons. The two groups of "particles" in the system, the electrons which interact via a short-range potential and the plasmons, were coupled to each other and among themselves individually be certain terms in the resulting Hamiltonian which could be considered small. Thus, in the random phase approximation for the plasmons (which simply means that plasmons of different momenta do not interact), the plasmon part of the Hamiltonian appears as a set of noninteracting harmonic oscillators, oscillating at the plasma frequency ω_0 . When the effect of the electron-plasmon coupling term was introduced (by means of a canonical transformation applied to the entire Hamiltonian of the system) the plasmon frequency became wavevector-dependent, and was given by

$$\omega_K^2 = \omega_0^2 + K^2\left(\frac{3}{5}v_0^2 - \frac{3}{20}\frac{\omega_0^2}{K_0^2}\right) + \frac{\hbar^2 K^4}{4m^2}$$

where $\omega_0^2 = 4\pi e^2 n_0/m$, v_0 is the Fermi velocity, $\hbar K_0$ is the Fermi momentum and n_0 is the density of electrons. This result is referred to throughout this thesis as the "usual microscopic-theory prediction".

Following closely on the heels of the Bohm-Pines work were the approaches of a number of other people using different techniques. Gell-Mann and Brueckner[2] showed that perturbation methods could be used from the outset (without

separating the Hamiltonian into collective and individual particle groups) if one would sum all the most-divergent terms in the perturbation series.

A number of workers have treated the problem by computing the dielectric response function for the electron gas. Using this approach, Nozieres and Pines[3] obtained a dispersion relation like the one above but without the term $-(3/20)\omega_0^2/k_0^2$. Their treatment was based on a self-consistent time-dependent perturbation calculation. By using diagram techniques, DuBois[4] computed the dielectric response function and from it determined the plasmon dispersion relationship, in agreement with that given above. Sawada et al[5] have used an equation-of-motion attack to compute the dielectric response function, and obtained a dispersion relation which agrees exactly with that of Nozieres and Pines. Ferrell[6] has used a self-consistent method modified to include exchange, obtaining a dispersion relation in agreement with that given above.

All of the above methods, while providing good descriptions of plasmons to lowest order in their various calculational procedures, suffer from the defect that attempts to include higher-order interactions always lead to burgeoning computational difficulties. The diagram techniques, and indeed any of the methods which involve infinite sums of

terms, are probably most celebrated in this respect. The various and numerous attempts to extend the diagram approach to other than the diagrams for lowest-order polarization (which is equivalent to the random phase approximation) and exchange, have led to results which are at best incomplete and at worst, simply wrong. (In the course of his work on this thesis the author has attempted both a self-consistent time-dependent approach and a diagram approach; the results of these efforts were both incomplete and wrong).

Probably the first paper to use hydrodynamics in treating the electron gas was that by Ritchie.[7] The hydrodynamic treatment was developed by Bloch[8] in the early thirties to study electrons in atoms. It treats the charged electron gas as a fluid which obeys the proper equations of motion for the gas. Wakano[9] was first to quantize the hydrodynamical model (in his work on atoms). Ritchie first used it in a form containing only the Pauli contribution to the pressure to predict the existence of surface plasma oscillations. The same model was used recently by Wilems and Ritchie[10] to incorporate the effects of both radiation and a beam of fast charged particles into the electron gas. They also used just the Pauli contribution to the pressure-density relation.

This thesis considers the problem of the consequences of plasmon-plasmon interactions within the electron gas.

The quantum hydrodynamic approach is used, together with the equation-of-motion technique, to include the effects of an expanded pressure-density relation, one which includes the Pauli term, the exchange contribution, and an expression which accounts for the correlations due to the Coulomb interactions.

The hydrodynamic treatment is conceptually similar to the Bohm-Pines collective coordinate approach. That is, the hydrodynamic attack recognizes implicitely that the collective motion is the result of the long-range part of the Coulomb interaction (reflected in the fact that the sums over wavevectors go only to some cutoff $k_c < k_f$, the Fermi momentum). It tacitly ignores the short-range interactions between the particles, i.e. interactions associated with wavevectors $k_c < k < k_f$.

Chapter II is devoted to deriving the proper Hamiltonian of the system to third order in the density fluctuations. The system is quantized and its relation to the microscopic theory is discussed.

Chapter III is concerned with obtaining the consequences of the nonlinear portion of the Hamiltonian, in the guise of a correction to the plasmon dispersion relation. An integral eigenvalue equation is obtained by the equation-of-motion method, and is solved to get the corrections to

to the plasma frequency.

Chapter IV examines the ramifications of the new plasma frequency for the problem of the electron lattice.

Chapter V briefly discusses the correction which the nonlinear terms add to the ground-state energy.

Chapter VI discusses possible experimentally-observable effects due to the nonlinear correction to the plasma dispersion relation. The available data from transmission exeriments are examined. Calculations of the contribution of the plasmon-plasmon interactions to the lifetime of a plasmon are carried out and discussed.

II. DERIVATION OF THE HAMILTONIAN

A. Equations of Motion and Lagrangian

1. Equations of Motion

In the hydrodynamic approach the electron gas must satisfy three equations of motion. The first is Poisson's equation:

$$\nabla^2\varphi = 4\pi e(n-n_0) \tag{2.1}$$

where φ is the electric potential and n is the number density of electrons. The second equation is the continuity equation:

$$-\frac{\partial n}{\partial t} = \nabla\cdot(n\vec{v})$$

here $\vec{v}$ is the velocity field. If there are no forces acting on the system which can cause the fluid to circulate (such as magnetic forces), we can set $\nabla\times\vec{v}=0$ and put $\vec{v}=-\nabla\psi$ where ψ is the so-called scalar velocity potential. The magnetic forces which arise from currents in the fluid can be neglected because they are of order v/c where c is the velocity of light. The continuity equation then becomes

$$\frac{\partial n}{\partial t} = \nabla\cdot(n\nabla\psi). \tag{2.2}$$

The dynamics are contained in the Euler equation

$$mn\frac{d\vec{v}}{dt}=-\nabla P+ne\nabla\varphi$$

where m is the mass of an electron, P is the pressure, and $\frac{d}{dt}=\frac{\partial}{\partial t}+\vec{v}\cdot\nabla$ is the co-moving time derivative. If we assume that P is a function of n, then $\nabla P=\frac{dP}{dn}\nabla n$. Substituting $\vec{v}=-\nabla\psi$ we have

$$\frac{d}{dt}\nabla\psi=\frac{1}{m}\frac{dP}{dn}\frac{\nabla n}{n}-\frac{e}{m}\nabla\varphi.$$

Applying the co-moving time derivative to $\nabla\psi$ we obtain

$$\frac{d}{dt}\nabla\psi=\nabla\left(\frac{\partial\psi}{\partial t}\right)-\frac{1}{2}\nabla(\nabla\psi)^2.$$

Therefore,

$$\nabla\frac{\partial\psi}{\partial t}=\frac{1}{2}\nabla(\nabla\psi)^2+\frac{1}{m}\frac{dP}{dn}-\frac{e}{m}\nabla\varphi.$$

We now form the scalar product of both sides of this equation with $\vec{dr}$ and use the fact that $dr\cdot\nabla\beta=d\beta$ where β is some scalar.[11] After integrating the resulting equation, we get the third equation of motion for the electron gas:

$$\frac{\partial\psi}{\partial t}=\frac{1}{2}(\nabla\psi)^2+\frac{1}{m}\int_0^n\frac{dP(n')}{n'}-\frac{e}{m}\varphi. \tag{2.3}$$

We now turn to the problem of determining the Lagrangian density which will give rise to Eqs. (2.1), (2.2) and (2.3).

2. The Lagrangian Density

As usual, the equations of motion follow from the Euler Lagrange equations which are obtained by varying the Lagrangian L such that

$$\delta \int_{t_1}^{t_2} L\, dt = 0$$

In terms of the Lagrangian density $\mathcal{L}$ these equations are

$$\frac{\partial}{\partial t} \frac{\partial \mathcal{L}}{\partial \left(\frac{\partial \Lambda}{\partial t}\right)} + \sum_{i=1}^{3} \frac{\partial}{\partial r_i} \frac{\partial \mathcal{L}}{\partial \left(\frac{\partial \Lambda}{\partial r_i}\right)} - \frac{\partial \mathcal{L}}{\partial \Lambda} = 0$$

where Λ is n, φ or ψ in turn. We shall show that the Lagrangian density which will produce the desired equations of motion is

$$\mathcal{L} = \frac{1}{8\pi}(\nabla\varphi)^2 + n\left[m\frac{\partial\psi}{\partial t} - \frac{m}{2}(\nabla\psi)^2 - \int_0^n \frac{P(n')dn'}{n'^2}\right] + e\varphi(n-n_0).$$

To show that this is a valid choice for $\mathcal{L}$, we first let $\Lambda = n$. The only term which appears is $\partial\mathcal{L}/\partial n$;

$$0 = m\frac{\partial\psi}{\partial t} - \frac{m}{2}(\nabla\psi)^2 + e\varphi - \frac{P(n)}{n} - \int_0^n \frac{P(n')dn'}{n'^2}.$$

The last two terms are just $\int_0^n \frac{dP(n')}{n'}$ integrated once by parts. Thus, this equation gives us Eq. (2.3).

Putting $\Lambda = \varphi$ we obtain

$$\frac{1}{4\pi}\nabla^2\varphi = e(n-n_0)$$

which is just Eq. (2.1). Finally, when $\Lambda = \Psi$ we get

$$\frac{\partial}{\partial t}(nm) - \nabla\cdot(nm\nabla\Psi) = 0 .$$

Upon dividing by m and putting $\nabla\Psi = -\vec{v}$ we find Eq. (2.2).

B. Hamiltonian

1. Classical Hamiltonian Density

The Hamiltonian is the volume integral of the Hamiltonian density which, in turn, is defined as

$$\mathcal{H} = \pi\frac{\partial\mathcal{L}}{\partial t} - \mathcal{L} \; ; \; \pi = \frac{\partial\mathcal{L}}{\partial\left(\frac{\partial\Psi}{\partial t}\right)} .$$

With our choice for $\mathcal{L}$ the momentum density π conjugate to Ψ is

$$\pi = mn \tag{2.4}$$

Thus, $\mathcal{H}$ becomes

$$\mathcal{H} = \frac{1}{2}mn(\nabla\Psi)^2 + n\int_0^n \frac{P(n')dn'}{n'^2} - e\varphi(n-n_0) - \frac{1}{8\pi}(\nabla\varphi)^2 . \tag{2.5}$$

Since we shall be interested in the volume integral of $\mathcal{H}$, let us examine the volume integral of the last term. By Green's theorem we have

$$-\frac{1}{8\pi}\int_V (\nabla\varphi)^2 dV = \frac{-1}{8\pi}\int_S \varphi\nabla\varphi\cdot d\vec{S} + \frac{1}{8\pi}\int_V \varphi\nabla^2\varphi\, dV .$$

We shall assume that φ goes to zero fast enough so that the surface integral will vanish. Replacing $\nabla^2\varphi$ with $4\pi e(n-n_0)$ in the resulting equation, we find that the Hamiltonian density is unaltered if we replace $-(1/8\pi)(\nabla\varphi)^2$ by $(1/2)e(n-n_0)$. Therefore, we shall take the Hamiltonian density to be

$$\mathcal{H}=\frac{1}{2}mn(\nabla\psi)^2+n\int_0^n\frac{P(n')dn'}{n'^2}-\frac{1}{2}e\varphi(n-n_0). \tag{2.6}$$

2. Reduction to Normal Coordinates

Our goal is to express $\mathcal{H}$ in a form containing a single field quantity, which we choose to be n. Therefore, ψ and φ must be expressed in terms of n; the equations of motion permit us to do so.

Let us first express all the fields in terms of Fourier series:

$$n(r,t)=\sum_q n_q(t)\,e^{iq\cdot r} \tag{2.7}$$

$$\varphi(r,t)=\sum_q \varphi_q(t)\,e^{iq\cdot r} \tag{2.8}$$

$$\psi(r,t)=\sum_q \psi_q(t)\,e^{iq\cdot r}. \tag{2.9}$$

Equation (2.1) allows us to relate φ to n. Thus

$$\varphi_q(t) = -\frac{4\pi e^2 n_q(t)}{q^2} . \tag{2.10}$$

We can use the continuity equation, Eq. (2.2), to relate n and ψ . After substituting the Fourier series into Eq. (2.2) we obtain the following relationship:

$$\dot{n}_q = -\sum_k n_{q-k} \psi_k k \cdot q .$$

With an approximation soon to be made in mind, we write this equation as

$$\dot{n}_q = -n_0 \psi_q q^2 - \sum_{k \neq q} n_{q-k} \psi_k k \cdot q .$$

Solving this equation for ψ_q and iterating the relationship thus obtained once, we have

$$\psi_q = -\frac{\dot{n}_q}{n_0 q^2} + \frac{1}{n_0^2 q^2} \sum_{k \neq q} n_{q-k} n_k \frac{k \cdot q}{k^2} . \tag{2.11}$$

The following assumption is now made. It will be assumed that the density n(r,t) can be written as

$$n(r,t) = n_0 + n_1(r,t)$$

where n_0 is independent of both time and position (it is the usual unperturbed charge distribution within the electron gas), and $n_1(r,t)$ contains all the deviation from equilibrium. Thus, n_0 is just the q=0 component of the Fourier series for

n(r,t) and is time independent. We therefore have

$$\frac{1}{2}mn(\nabla\psi)^2-\frac{1}{2}\phi e(n-n_0)=\frac{1}{2}(n_0+n_1)m(\nabla\psi)^2-\frac{1}{2}e\varphi n_1 .$$

Upon substituting the Fourier series into this equation and performing the volume integration required, we have

$$\left[\frac{m}{2}n_0\sum_k k^2\psi_k\psi_{-k}-\frac{m}{2}\sum_{kk'}\psi_k\psi_{k'}n_{-k-k'}k\cdot k'+\frac{4\pi e^2}{2}\sum_k\frac{1}{k^2}n_k n_{-k}\right]V.$$

The above is the total Hamiltonian for the system, without the pressure term which does not involve ψ (we shall treat the pressure term in the same manner presently). In terms of the conjugate variables π_k and ψ_k, this part of the Hamiltonian is

$$H'=\left[\frac{m}{2}n_0\sum_k k^2\psi_k\psi_{-k}-\frac{1}{2}\sum_{kk'}\psi_k\psi_{k'}\pi_{-k-k'}k\cdot k'+\frac{4\pi e^2}{2m^2}\sum_k\frac{1}{k^2}\pi_k\pi_{-k}\right]V.$$

Because the portion of H tied up in the pressure term involves only n, it can be expressed solely in terms of π. We now proceed to drop the kinetic energy term which contains $\psi_k\psi_{k'}\pi_{-k-k'}$. This is done for two reasons. The first is that this term will destroy the symmetry needed to perform subsequent calculations when solving the equation of motion. Second, we intend to seek corrections to the plasmon energy which are valid to second order in the wavevector; this kinetic energy term will introduce terms of higher order than k^2. H' is then

$$H' = \frac{1}{2}\sum_{k}\left(m n_0 k^2 \psi_k \psi_{-k} + \frac{\omega_0^2}{m n_0 k^2}\pi_k \pi_{-k}\right)V. \qquad (2.12)$$

We have put in the classical plasma frequency $\omega_0^2 = 4\pi e^2 n_0/m$. It is clear that with a judicious redefinition of ψ_k and π_k H' can be recast in the usual harmonic oscillator form, with frequency ω_0.

C. The Pressure Terms

We shall now consider the pressure-density term which has been neglected until now. Recall that the pressure part of the Hamiltonian is written as

$$H_P = \int dV\, n \int_0^n \frac{P(n')\,dn'}{n'^2}.$$

To evaluate this contribution we need to know how the pressure depends on the density for our particular problem. The pressure-density relation we shall use is of the form

$$P(n) = P_1(n) + P_2(n) + P_3(n) + P_4(n)$$

where

$$P_1(n) = \frac{h^2}{5m}\left(\frac{3}{8\pi}\right)^{2/3} n^{5/3} \qquad (2.12a)$$

$$P_2(n) = -\frac{1}{2}e^2\left(\frac{3}{8\pi}\right)^{1/3} n^{4/3} \qquad (2.12b)$$

$$P_3(n) = -\frac{1}{3}\frac{abn^{4/3}}{(n^{1/3}+b)^2} \tag{2.12c}$$

$$a = \frac{0.05647e^2}{a_o}, \quad b = \frac{0.793}{a_o}, \quad a_o = \frac{\hbar^2}{e^2 m}$$

$$P_4(n) = n^2 \frac{d}{dn}\left(\frac{\nabla n}{n}\right)^2 \frac{1}{8} e^2 a_o . \tag{2.12d}$$

$P_1(n)$ gives rise to the well-known "Pauli Exclusion Principle" energy, which is the kinetic energy of a collection of Fermions at temperature T=0. The second term, $P_2(n)$, is that obtained by Dirac for taking account of the exchange energy of electrons with parallel spins. $P_3(n)$ was obtained by Wigner and accounts for the correlation between electrons with antiparallel spins. The fourth term, $P_4(n)$, comes from the so-called Weizsäcker inhomogeneity correction. The Weizsäcker term essentially accounts for the kinetic energy at T=0 if the density is not uniform (a derivation of this term is presented in Appendix B). Its contribution to the Hamiltonian takes the form

$$H = \int dV \frac{1}{8} e^2 a_o \frac{(\nabla n)^2}{n} \tag{2.13}$$

where a_o is the Bohr radius. References for all these pressure terms are given in Gombas.[12]

Now

$$n\int_0^n \frac{P_1(n')}{n'^2}dn' = \frac{3}{2}\frac{h^2}{5m}\left(\frac{3}{8\pi}\right)^{2/3} n^{5/3}, \tag{2.14a}$$

$$n\int_0^n \frac{P_2(n')}{n'^2}dn' = \frac{-3}{2}e^2\left(\frac{3}{8\pi}\right)^{1/3} n^{4/3}, \tag{2.14b}$$

$$n\int_0^n \frac{P_3(n')}{n'^2}dn' = \frac{-a n^{4/3}}{n^{1/3}+b}, \tag{2.14c}$$

$$n\int_0^n \frac{P_4(n')}{n'^2}dn' = \frac{1}{8}e^2 a_0\left(\frac{\nabla n}{n}\right)^2. \tag{2.14d}$$

We shall now expand each of the above terms to third order in $n_1(r,t)$.

1. <u>Pauli Term</u>

We set all the constants in Eq. (2.14a) equal to A for the moment and consider

$$An^{5/3} = An_0^{5/3}\left(1+\frac{5}{3}\frac{n_1}{n_0}+\frac{5}{9}\left(\frac{n_1}{n_0}\right)^2-\frac{5}{81}\left(\frac{n_1}{n_0}\right)^3\right).$$

In the volume integral which gives us the Hamiltonian from the pressure, the second term above (proportional to n_1) drops out because of conservation of matter. The lowest-order term contributes a constant, and the total energy from the Pauli term is

$$E_1 = E_0' + An_0^{5/3}\int dv\left[\frac{5}{9}\left(\frac{n_1}{n_0}\right)^2-\frac{5}{81}\left(\frac{n_1}{n_0}\right)^3\right],\quad E_0' = An_0^{5/3}V. \tag{2.15a}$$

We shall drop the constant energy term for the moment since it will not influence the dynamics of the electron gas. After substituting the Fourier expansion of n_1 into Eq. (2.15a) and converting to momentum density components we have

$$E_1 = An_0^{5/3}\left[\sum_k \frac{5}{9}\frac{1}{(mn_0)^2}\pi_k\pi_{-k} - \frac{5}{81}\frac{1}{(mn_0)^3}\sum_{kq}\pi_k\pi_q\pi_{-k-q}\right]V. \quad (2.15b)$$

The quadratic term above can be combined with the quadratic part of the harmonic oscillator Hamiltonian mentioned above, producing a change in the oscillator frequency.

2. Exchange Term

Collecting the constants in Eq. (2.14b) into B, we have an expression of the form

$$Bn^{4/3} = Bn_0^{4/3}\left(1+\frac{4}{3}\frac{n_1}{n_0}+\frac{2}{9}\left(\frac{n_1}{n_0}\right)^2-\frac{4}{81}\left(\frac{n_1}{n_0}\right)^3\right).$$

Again, after Fourier transforming and changing to we get

$$E_2 = Bn_0^{4/3}\left[\frac{2}{9}\frac{1}{(mn_0)^2}\sum_k \pi_k\pi_{-k} - \frac{4}{81}\frac{1}{(mn_0)^3}\sum_{kq}\pi_k\pi_q\pi_{-k-q}\right]V. \quad (2.15c)$$

We have dropped the unperturbed energy $E_2^0 = Bn_0^{4/3}V$. This expansion will also contribute to a change in the oscillator frequency.

3. Correlation Term

Equation (2.14c) gives the following Hamiltonian density when expanded to third order

$$\mathcal{H}_3 = a n_0 c\left[1+\frac{\Delta}{3}(4-c)+\frac{\Delta^2}{9}(c^2-3c+2)+\frac{\Delta^3}{81}(-3c^3+6c^2+c-4)\right];$$

$$c=\frac{n_0^{1/3}}{n_0^{1/3}+b}, \quad \Delta=\frac{n_1}{n_0}.$$

When integrated over the volume, the term linear in Δ is zero and we obtain a constant energy of

$$E_3^0 = -a n_0 V \frac{n_0^{1/3}}{n_0^{1/3}+b}.$$

Once again Fourier transforming and converting to momentum density components, we obtain

$$E_3 = -\frac{a n_0^{1/3}}{n_0^{1/3}+b}\left[\frac{c^2-3c+2}{9}\sum_k \frac{1}{(m n_0)^2}\pi_k \pi_{-k} + \frac{-3c^3+6c^2+c-4}{81}\sum_{kk'}\frac{1}{(m n_0)^3}\pi_k \pi_{k'} \pi_{-k-k'}\right]V. \qquad (2.15d)$$

4. Weizsacker Term

From Eq. (2.13) we find

$$E_4 \approx \frac{1}{8} e^2 a_0 \int dV \frac{(\nabla n_1)^2}{n_0}\left(1-\frac{n_1}{n_0}\right)$$

$$= \frac{e^2 a_0}{8 n_0}\left[\sum_k k^2 \frac{1}{m^2}\pi_k \pi_{-k} - \frac{1}{n_0 m^3}\sum_{kk'} k\cdot k' \pi_k \pi_{k'} \pi_{-k-k'}\right]V. \qquad (2.15e)$$

Note that this term introduces no constant energy.

D. Quantization

Let us now combine Eq. (2.12) with the quadratic parts of Eqs. (2.15b-e):

$$H_2 = \frac{1}{2}\sum_k \left(m n_0 k^2 \psi_k \psi_{-k} + \frac{\omega_k^2}{k^2 m n_0} \pi_k \pi_{-k}\right) V; \tag{2.16}$$

$$\omega_k^2 = \omega_0^2 + k^2 \left[\left(\frac{3}{8\pi}\right)^{2/3} \frac{\hbar^2 n_0^{2/3}}{3m^2} - \frac{e^2}{3}\left(\frac{3}{\pi}\right)^{1/3} \frac{n_0^{1/3}}{m} - \frac{c^2 - 3c + 2}{9m} \frac{a n_0^{1/3}}{n_0^{1/3} + b}\right] + k^4 \frac{e^2 a_0}{4m}. \tag{2.17}$$

We now quantize Eq. (2.16) in the usual manner by demanding that the commutators of the conjugate variables satisfy these equations:

$$[\pi_k, \psi_{k'}] = -i\hbar\,\delta_{k,k'}; \quad [\pi_k, \pi_{k'}] = [\psi_k, \psi_{k'}] = 0.$$

The required transformations are

$$\begin{aligned} \psi_k &= -\frac{i}{k}\left[\frac{\hbar\omega_k}{2 m n_0 V}\right]^{1/2} (b_k^+ - b_{-k}) \\ \pi_k &= k\left[\frac{\hbar m n_0}{2\omega_k V}\right]^{1/2} (b_k + b_{-k}^+), \end{aligned} \tag{2.18}$$

where $[b_k, b_{k'}^+] = \delta_{k,k'}$, $[b_k, b_{k'}] = [b_k^+, b_{k'}^+] = 0$. With these transformations Eq. (2.16) reduces to the celebrated harmonic oscillator form:

$$H_2 = \sum_k \hbar\omega_k \left(b_k^+ b_k + \frac{1}{2}\right).$$

Let us now examine the oscillator frequency as given in

Eq. (2.17). The first term is the usual classical plasma frequency. The second term can be reduced to a more transparent form by making use of the defining equations for the Fermi wavevector k_0 ($n_0 = (3\pi^2)^{-1} k_0^3$) and velocity $v_0 = \hbar k_0/m$; it becomes

$$\frac{1}{3} k^2 v_0^2 .$$

This result is the same as that obtained by Wilems and Ritchie[10] and compares favorably with the usual result produced by the microscopic theories (which is $(3/5)k^2 v_0^2$).

The third term in Eq. (2.17) reduces to

$$-\frac{\omega_0^2}{4} \frac{k^2}{k_0^2} .$$

This contribution corresponds to the correction produced in the usual microscopic theory due to the coupling between electrons and plasmons which is outside the scope of the random phase approximation (RPA).[13] In the microscopic theory this correction amounts to $-(3/20)\omega_0^2 k^2/k_0^2$. Our correction is 5/3 this quantity. The hydrodynamic approach shows that this correction is in fact one partly resulting from correlation between electrons with parallel spins.

The fourth term in Eq. (2.17) is

$$-\frac{0.05647 e^2}{a_0} \frac{k^2}{m} \frac{c^3 - 3c^2 + 2c}{9} , \quad c = \frac{n_0^{1/3}}{n_0^{1/3} + b} .$$

It results from the correlation of electrons with antiparallel spins, and has not yet been produced in a microscopic theory. Its density dependence is interesting; as the density n_0 becomes very large, the quantity c goes to one. The combination c^2-3c+2 then goes to zero, causing the entire term to go to zero. This is, of course, what one would expect; the contribution due to correlation of electrons with antiparallel spins should become small as the density increases. Note too that the presence of this term does not contradict the result obtained from the usual microscopic theory that the RPA is exact in the high-density limit.

Finally, let us examine the last term in Eq. (2.17). It reduces to

$$\frac{\hbar^2 k^4}{4m^2}$$

which is exactly that produced by the microscopic theory. But it represents more than just agreement with the usual microscopic theory; it is also a confirmation of the validity of the Weizsäcker contribution to the hydrodynamic model. Confirmations of the Weizsäcker contribution to any problem are hard to come by.[12]

To complete our discussion of the Hamiltonian, we shall briefly discuss the energy per particle in the ground state. This quantity is available to us from the various constant

energies which have been calculated and systematically ignored until now. Collecting E_o^1, E_o^2, and E_o^3 from Secs. 1, 2, and 3, adding and dividing by N (the number of electrons in the gas), we find the energy per particle to be

$$\mathcal{E}_o = \frac{3}{2}\frac{h^2}{5m}\left(\frac{3}{8\pi}\right)^{2/3} n_o^{2/3} - \frac{3}{2}e^2\left(\frac{3}{8\pi}\right)^{1/3} n_o^{1/3} - \frac{0.05647e^2}{a_o}\frac{n_o^{1/3}}{n_o^{1/3}+\frac{0.793}{a_o}}.$$

This can be expressed in the more common form if we write n_o in terms of the radius per particle, r_o, and the dimensionless density parameter $r_s = r_o/a_o$:

$$\mathcal{E}_o = \frac{2.21}{r_s^2} - \frac{0.916}{r_s} - \frac{0.88}{r_s+7.8}\,Ry. \tag{2.19}$$

The last term above represents Wigner's calculation of the correlation energy; it is designed to be approximately valid at both high and low densities.

III. PLASMON-PLASMON INTERACTION

We shall pursue the following program. The Hamiltonian derived in Chapter II contains terms valid through third order in the creation and annihilation operators for density fluctuations. When expressed in harmonic oscillator form, these density fluctuations are referred to as plasmons, and the frequency of a plasmon with a given wavevector is given in Eq. (2.17). The third-order term in the Hamiltonian describes the interactions of plasmons with plasmons; it is very much analogous to the term encountered when dealing with phonon-phonon interactions. Because of the interaction the frequency, and therefore the energy, of a plasmon will be altered from that for the non-interacting "bare" plasmon, as specified in Eq. (2.17). We shall use the equation-of-motion method to determine the energy of the "physical" plasmon.

A. Equation of Motion

The third-order contribution to the Hamiltonian is contained in Eqs. (2.15b-d). The third-order term from the Weizsacker contribution will not be retained because it will produce corrections to the frequency which are higher order

than k^2, and we shall treat only k^2 corrections. Upon quantizing the third-order contributions and performing some algebra, we find the new correction to be

$$H_3 = \sum_{kq} W(k,q)(b_k + b_{-k}^+)(b_q + b_{-q}^+)(b_{-k-q} + b_{k+q}^+),$$

where

$$W(k,q) = |k||q||k+q| \left[\frac{\hbar^3 m^3 n_0^3}{8V\omega_k \omega_q \omega_{k+q}}\right]^{1/2} \left[-\frac{h^2}{54m^4}\left(\frac{3}{8\pi}\right)^{2/3} n_0^{-4/3} + \left(\frac{3}{\pi}\right)^{1/3} \frac{e^2}{27m^3 n_0^{5/3}} - \frac{a}{m^3 n_0^2 81}(-3c^4 + 6c^3 + 11c^2 - 4c)\right]. \tag{3.1}$$

The total Hamiltonian for the system is then

$$H = \sum_k \hbar\omega_k b_k^+ b_k + \sum_{kq} W(k,q)[b_k b_q b_{-k-q} + 3b_{k+q}^+ b_k b_q + 3b_k^+ b_q^+ b_{k+q} + b_k^+ b_q^+ b_{-k-q}^+]. \tag{3.2}$$

We have employed the commutation relations for the b_k to obtain this form, and all the terms linear in b_k or b_k^+ have been dropped (they enter with coefficients which are zero either because of attendant delta functions or because of the form of W(k,q)). Explicit use has been made of the symmetry of ω_k in order to cast H in the form above.

The equation of motion method tells us to seek operators O_k^+ which satisfy the equation

$$[H, O_k^+] = E_k O_k^+. \tag{3.3}$$

(See Nordtvedt[14] for a detailed discussion of the method). The energy E_k is the excitation energy; it is not the absolute energy for a given state.

Following Nordtvedt, we now postulate the form for $O_{k'}^{+}$ to be a linear combination of bare plasmon operators containing both single operators and quadratic combinations of operators, all chosen to create an excitation of momentum k' in the system; that is,

$$O_{k'}^{+} = \alpha b_{k'}^{+} + \beta b_{-k'} + \sum_{k''} \left(\gamma_{k''} b_{k''}^{+} b_{k'-k''}^{+} + \gamma_{k''}' b_{-k''} b_{-k'+k''} + \gamma_{k''}'' b_{k''+k'}^{+} b_{k''} \right). \tag{3.4}$$

Our choice of $O_{k'}^{+}$ can be viewed in a sense as an attempt to factor the new Hamiltonian so that it might be written in the usual harmonic oscillator form, but now in terms of the new operators O_k and O_k^{+}. The coefficients $\alpha, \beta, \gamma_{k''}, \gamma_{k''}', \gamma_{k''}''$ in Eq. (3.4) are to be determined.

Substituting this expression into Eq. (3.3) and calculating the necessary commutators (which are collected in Appendix A), we find $[H, O_{k'}^{+}]$ to be

$$\sum_{k} \hbar\omega_k \Big[\alpha b_k^{+} \delta_{k,k'} - \beta b_k \delta_{k,-k'} + \sum_{k''} \gamma_{k''} \left(b_k^{+} b_{k'-k''}^{+} \delta_{k,k''} + b_{k''}^{+} b_k^{+} \delta_{k,k'-k''} \right) + \sum_{k''} \gamma_{k''}' \left(-b_k b_{-k'+k''} \delta_{-k'',k} - b_{-k''} b_k \delta_{k,-k'+k''} \right)$$

$$+\sum_{k''}\gamma''_{k''}\left(b^{+}_{k}b_{k''}\delta_{k,k''+k'}-b^{+}_{k''+k'}b_{k}\delta_{k,k''}\right)\Big]+\sum_{kq}W(k,q)\Big\{3\alpha\times$$

$$b_{q}b_{-q-k}\delta_{k',k}+6\alpha b^{+}_{k+q}b_{q}\delta_{k,k'}+3\alpha b^{+}_{k}b^{+}_{q}\delta_{k',k+q}$$

$$-3\beta b_{k}b_{q}\delta_{k+q,-k'}-6\beta b^{+}_{q}b_{k+q}\delta_{k,-k'}-3\beta b^{+}_{q}b^{+}_{-k-q}\delta_{k,-k'}$$

$$-3\beta b_{k}b_{q}\delta_{k+q,-k'}-6\beta b^{+}_{q}b_{k+q}\delta_{k,-k'}-3\beta b^{+}_{q}b^{+}_{-k-q}\delta_{k,-k'}$$

$$+\sum_{k''}\Big(3\gamma_{k''}\left(b_{k}b_{-k-q}b^{+}_{k'-k''}\delta_{k'',q}+b^{+}_{k''}b_{k}b_{-k-q}\delta_{k'-k'',q}\right)$$

$$+6\gamma_{k''}\left(b^{+}_{k+q}b_{k}b^{+}_{k'-k''}\delta_{k'',q}+b^{+}_{k''}b^{+}_{k+q}b_{k}\delta_{q,k'-k''}\right)$$

$$-6\gamma'_{k''}\left(b^{+}_{k}b_{k+q}b_{-k'+k''}\delta_{q,-k''}+b_{-k''}b^{+}_{k}b_{k+q}\delta_{q,-k'+k''}\right)$$

$$-3\gamma'_{k''}\left(b^{+}_{k}b^{+}_{q}b_{-k'+k''}\delta_{k'',k+q}+b_{-k''}b^{+}_{k}b^{+}_{q}\delta_{-k'+k'',-k-q}\right)$$

$$+3\gamma''_{k''}\left(2b^{+}_{k+q}b_{k}b_{k''}\delta_{q,k''+k'}-b^{+}_{k''+k'}b_{k}b_{q}\delta_{k+q,k''}\right)$$

$$+3\gamma''_{k''}\left(b^{+}_{k}b^{+}_{q}b_{k''}\delta_{k+q,k'+k''}-2b^{+}_{k''+k'}b^{+}_{k}b_{k+q}\delta_{q,k''}\right)\Big\}.$$

Using the delta functions in the above expression, we perform the sums over the wavevectors. The terms containing three operators are treated as follows. After performing the initial sum over wavevectors, a typical triplet is

$$\sum_{kq}W(k,q)\,3\gamma_{q}b_{k}b_{-k-q}b^{+}_{k'-q}\,.$$

The commutation rules are applied once to obtain

$$\sum_q 3W(-k';q)\,\gamma_q\, b_k + \sum_{kq} 3W(k,q)\,\gamma_q\, b_k\, b^+_{k'-q}\, b_{-k-q}\,.$$

The triplet in the above expression somewhat resembles a single operator b_k times a number operator (it is not a true number operator, of course, because the indices are mixed). When the operators in the triplet are replaced by their ground-state expectation values, the triplet goes to zero, leaving us with just the first term containing b_k. After treating all the triplet terms in this manner, the entire sum is rearranged, using the symmetry of W(k,q) whenever required, to give this sum:

$$b^+_{k'}\Big[\alpha\hbar\omega_{k'} + 6\sum_q W(k';-q)(\gamma_q-\gamma'_q)\Big] + b_{-k'}\Big[-\beta\hbar\omega_{k'} + 6\sum_q W(k';-q)(\gamma_q-\gamma'_q)\Big] + \sum_{k''} b^+_{k''}b^+_{k'-k''}\Big[\hbar(\omega_{k''}+\omega_{k'-k''})\gamma_{k''} + 3(\alpha-\beta)W(-k';k'')\Big] + \sum_{k''} b_{-k''}b_{k''-k'}\Big[-\hbar(\omega_{k''}+\omega_{k'-k''})\gamma'_{k''} + 3(\alpha-\beta)W(k';-k'')\Big] + \sum_{k''} b^+_{k''+k'}b_{k''}\Big[\hbar(\omega_{k''+k'}-\omega_{k''})\gamma''_{k''} + 6(\alpha-\beta)W(k';k'')\Big].$$

The above expression is the commutator of H with the operator $O^+_{k'}$. Equating the coefficients of the operators in it (after changing k' to k for convenience) with those found

by multiplying Eq. (3.4) by E_k, we obtain the following series of equations:

$$\alpha E_k = \alpha \hbar \omega_k + 6 \sum_q W(k,-q)(\gamma_q - \gamma_q') \tag{3.5a}$$

$$\beta E_k = -\beta \hbar \omega_k + 6 \sum_q W(k,-q)(\gamma_q - \gamma_q') \tag{3.5b}$$

$$\gamma_{k'} E_k = \gamma_{k'}(\omega_{k'} + \omega_{k-k'})\hbar + 3(\alpha-\beta) W(-k,k') \tag{3.5c}$$

$$\gamma_{k'}' E_k = -\gamma_{k'}' \hbar(\omega_{k'} + \omega_{k-k'}) + 3(\alpha-\beta) W(k,-k') \tag{3.5d}$$

$$\gamma_{k'}'' E_k = \gamma_{k'}'' \hbar(\omega_{k'+k} - \omega_{k'}) + 6(\alpha-\beta) W(k,k'). \tag{3.5e}$$

B. Corrected Plasmon Energy

We shall now solve Eq. (3.5) to get an expression for the corrected plasmon energy, E_k. First, add and subtract Eqs. (3.5a) and (3.5b) and solve for $\alpha-\beta$:

$$\alpha-\beta = \frac{12\hbar\omega_k \sum_q W(k,-q)(\gamma_q - \gamma_q')}{E_k^2 - \hbar^2\omega_k^2}. \tag{3.6}$$

Solving for γ_q and γ_q' from Eqs. (3.5c) and (3.5d) we have

$$\gamma_q = \frac{3(\alpha-\beta)W(k,-q)}{E_k - \hbar(\omega_q + \omega_{k-q})}\ ;\quad \gamma_q' = \frac{3(\alpha-\beta)W(k,-q)}{E_k + \hbar(\omega_q + \omega_{k-q})}. \tag{3.7}$$

Finally, forming the sum present in Eq. (3.6), we obtain the following eigenvalue equation:

$$1 = \frac{72\hbar\omega_k}{E_k^2 - \hbar^2\omega_k^2} \sum_q \frac{\hbar(\omega_q + \omega_{k-q}) W^2(k,-q)}{E_k^2 - \hbar^2(\omega_q + \omega_{k-q})^2} . \tag{3.8}$$

This defines E_k^2 and is the fundamental result of this thesis.

This eigenvalue equation has a number of solutions, one which corresponds to the single plasmon operators in O_k^+ and another which corresponds to the two creation and two annihilation operators. In addition, if the operator O_K^+ is properly normalized to begin with, a third solution for E_k can be obtained from Eq. (3.5e). We will solve the integral equation, Eq. (3.8), by iteration. Thus, the E_k under the summation is replaced by $\hbar\omega_k$. Then, keeping terms such that the entire correction to $\hbar\omega_k$ is of order k^2 (which amounts to replacing all ω_k by ω_o under the summation), we find

$$E_k^2 = \hbar^2\omega_k^2 - 48 \sum_q W^2(k,-q). \tag{3.9}$$

Taking W(k,q) from Eq. (3.1) and changing ω_k to ω_o in it, transforming the sum in Eq. (3.9) to an integral over a sphere of radius k_c, and keeping only terms of order k^2, we have that

$$\sum_q W(k,-q) \rightarrow \frac{V}{(2\pi)^3} A^2 k^2 \int_0^{k_c} q^4 d^3q .$$

Here A contains all the momentum-independent parts of W(k,q). k_c is the cutoff momentum for plasmons, as yet unspecified. Therefore,

$$E_k^2 = \hbar^2\omega_k^2 - \frac{24V}{7\pi^2} A^2 k^2 k_c^7, \tag{3.10}$$

where

$$A^2 = \left[\frac{\hbar^3 m^3 n_o^3}{8\omega_o^3 V}\right]\left[-\frac{\hbar^2}{54m^4}\left(\frac{3}{8\pi}\right)^{2/3} n_o^{-4/3} + \left(\frac{3}{\pi}\right)^{1/3}\frac{e^2}{27m^3 n_o^{5/3}} - \frac{ac'}{m^3 n_o^2 81}\right]; \quad c' = -3c^4 + 6c^3 + c^2 - 4c, \tag{3.11}$$

and ω_k^2 is given by Eq. (2.17).

Let us now express E_k^2 in terms of the conventional notation using r_s, the dimensionless density parameter. r_s is defined as r_o/a_o, the radius per particle divided by the Bohr radius a_o. In terms of r_s, n_o is

$$n_o^{1/3} = \frac{\beta_o}{r_o}, \quad \beta_o = \left(\frac{3}{4\pi}\right)^{1/3}.$$

Substituting this into Eqs. (3.11) and (2.17) and doing some algebra, we obtain

$$E_k^2 = \frac{12}{r_s^3} + (a_o k)^2\left[\frac{4.88}{r_s^2} - \frac{0.815}{r_s} - 0.0251(c^3 - 3c^2 + 2c) - 0.002295(a_o k_c)^7 r_s^{7/2}(-0.629 + 0.175 r_s - 0.0054 r_s^2 c')^2\right] Ry^2 \tag{3.12}$$

in Rydbergs squared. The first term is the classical plasmon energy squared, the first three terms multiplied by k^2

are the Pauli, exchange, and correlation contributions obtained in the usual linear theory, and the last three terms, which are squared, are the nonlinear Pauli, exchange, and correlation contributions.

The high-density limit corresponds to small r_s and the low-density limit to large r_s. Therefore, as the density gets very high, the linear Pauli term represents the largest correction to the classical plasmon energy (as mentioned previously, the linear correlation energy goes to zero). This behavior confirms that the RPA is indeed the correct solution at high densities (we are implicitly assuming small momentum).

The linear Pauli and exchange terms cancel when the density parameter r_s=6.00. All three linear terms cancel at a slightly lower value of r_s. The value of r_s for real metals lies between 2 and 5.5; therefore one would not expect to see experimental evidence of the above cancelation. The r_s at which the entire k^2 coefficient is zero is altered only slightly when the nonlinear corrections are included.

The problem of specifying the cutoff k_c must now be faced. The cutoff is determined when the plasmon wavevector reaches a value at which the plasmon can decay and excite an electron out of the Fermi sea. Conservation of energy then dictates that

$$\hbar\omega_{k_c} = \frac{\hbar^2(k_c+k_f)^2}{2m} - \frac{\hbar^2 k_f^2}{2m}$$

where we have picked an electron at the Fermi surface to be excited. The lowest possible k_c will be determined when k_c and k_f are parallel. Keeping only the terms linear in k_c, we have

$$\hbar\omega_o = \frac{k_c}{k_f} 2E_f ,$$

or

$$\beta = 0.471\sqrt{r_s} \tag{3.13}$$

where $\beta = k_c/k_f$. More exact calculations of β (see Sawada et al[5] and Ferrell[6]) give a relationship between r_s and β which is very close to Eq. (3.13) at high and metallic densities ($r_s < 6$). Thus, we feel confident that Eq. (3.13) is valid in that region of densities.

Because we shall eventually want to consider the low-density region, we must try to estimate the validity of Eq. (3.13) in that region. With future developments in mind, consider an electron lattice with a body-centered cubic cell. If the length of one side of the cube is L, and there is one electron at each site, then the density is related to the volume per particle as

$$\frac{2}{L^3} = \frac{1}{\frac{4\pi}{3} r_0^3} .$$

The smallest possible wavelength in such a lattice is of the order L. Therefore, with $k_c^3 = (2\pi/L)^3 = 3\pi^2/r_0^3$ and $k_f = 1.92/r_0$, we find β to be

$$\beta = 1.61 .$$

Ferrell's expression for β gives a value $r_s = 17.1$ at $\beta = 1.61$; Eq. (3.13) gives $r_s = 11.7$ for the same β , which is about 30% lower than the Ferrell value. In spite of this difference, we shall use Eq. (3.13) because of its analytic simplicity. Results derived with it in this region of densities will be quantitatively unreliable, but the qualitative features should be unaltered.

Thus, taking $\beta = 0.471\sqrt{r_s}$, we arrive at our equation for the plasmon energy:

$$E_k^2 = \frac{12}{r_s^3} + (a_0 k)^2 \Big[\frac{4.88}{r_s^2} - \frac{0.815}{r_s} - 0.0251(c^3 - 3c^2 + 2c) - 0.00114(-0.529 + 0.175 r_s - 0.0054 r_s^2 c')^2 \Big] Ry^2 . \tag{3.14}$$

IV. THE ELECTRON LATTICE

Some thirty years ago, based on studies of the electron gas distributed in a uniform background of positive charge, Wigner[15] stated that as the density of the gas drops, the gas will "crystallize" into a body-centered cubic lattice. Calculations by Fuchs[16] using the Ewald technique indicate that for a given density the bcc lattice has the lowest energy for a stationary lattice; a face-centered cubic is next in line. Carr[17] states that he has performed a similar calculation for a simple cubic lattice and found its energy to be higher than either the bcc or fcc lattices. Kohn and Schechter[18] report calculations for a hexagonal close-packed structure. All of these lattices produce energies per particle which are very close to one another, but with the bcc having the lowest energy by a very narrow margin.

Pines[19] gives a review of the electron lattice from the Wigner-Seitz cell point of view. Basically, the electron lattice becomes stable at low densities because the potential energy, which favors the formation of a lattice, varies as $1/r_s$, while the kinetic energy, which militates against a lattice structure, goes as $1/r_s^2$. At low densities (high r_s) the potential term is dominant.

The question concerning the density at which the electron gas goes into the lattice has received renewed attention of late. By approaching the question from the point of view of determining the density at which the lattice would melt as one goes from low densities to high densities (using the Lindeman melting criterion) Nozieres and Pines[20] conclude that the lattice would become unstable for $r_s \lesssim 20$.

Carr[17] has examined the electron lattice with a perturbation technique, taking into account the zero-point oscillations. He found that there was no breakdown in his perturbation expansion for r_s greater than about 5. Coldwell-Horsfall and Maradudin[21], in their treatment of the electron lattice, have determined the lattice to be stable for $r_s=6.5$. Their result was derived from the Lindeman melting criterion as was that of Nozieres and Pines.

A more recent study of the problem by de Wette[22] produces a different estimate. Based on the notion that a solid structure will exist when the single particle potential exhibits at least one bound state, he determines the density at which the bound state disappears. This corresponds to melting; he finds that the upper and lower limits on the melting region are given by $47 \leqslant r_s \leqslant 100$.

We can attack this problem from still a different point of view with the results derived in the last chapter. The

essential point of this approach is that plasmon oscillations are density fluctuations. If, at some density, the time dependence of the oscillations changes from sinusoidal to purely exponential, we are faced with a phase transition in which the density fluctuations appear to grow in time. At this juncture the model ceases to be viable; we can depend on it only to signal the transition, but it will not describe the new state.

Therefore, consider the equation of motion which we used earlier:

$$[H, O_k^+] = E_k O_k^+ = -i\hbar \dot{O}_k^+$$

We then have that $\dot{O}_k^+ = (i/\hbar) E_k O_k^+$, or that

$$\ddot{O}_k^+ + \frac{E_k^2}{\hbar^2} O_k = 0.$$

The solutions to this differential equation are of the form

$$A e^{i\frac{E_k}{\hbar}t}.$$

Since the O_k^+ are operators which create plasmons (i.e. density fluctuations) we see that if E_k becomes imaginary we have the possibility of creating density fluctuations which grow in time, i.e. a phase transition. We may therefore gain some insight into the electron lattice by examining the behavior of E_k as a function of density.

Calculations of E_k^2 have been performed using Eq. (3.14) in which k has been selected such that it is very nearly the wavevector corresponding to the shortest wavelength possible in a bcc lattice (that wavelength is the distance from a corner electron to the electron in the center). The wavevector selected also ensures that the number of plasmon modes does not exceed the total number of modes in the system. That is, since the number of plasmon modes is $n=(4\pi/3)k^3(2\pi)^{-3}$ and the total number of modes is $3n_0=(8\pi/3)(2\pi)^{-3}k_0^3$ (with the volume V=1), we have

$$\frac{n}{3n_0}=\frac{1}{6}\left(\frac{k}{k_0}\right)^3.$$

This ratio is 1 when $k=3.49/r_0$. With this k we have found that

$$E_k^2=0 \text{ for } r_s\approx 16.9. \tag{4.1}$$

This is not an unreasonable value for r_s at the transition point, but in view of the approximations used one cannot depend on it too heavily. It appears feasible that the model will indicate a phase transition, but the exact density at which it occurs depends rather strongly on the approximations employed.

Various portions of the expression for E_k^2 have been plotted. Figure 1 gives the various contributions from the

"quadratic" part of E_k^2. These contributions and their curves are:

$$\frac{4.88}{r_s^2} \quad \text{curve 1}$$

$$\frac{0.815}{r_s} \quad \text{curve 2}$$

$$0.0251(c^3-3c^2+2c) \quad \text{curve 3.}$$

As mentioned earlier, the correlation term (Curve 3) is zero when $r_s=0$. The exchange curve (2) crosses the Pauli curve (1) at $r_s=6$. The Pauli term is canceled by the sum of the exchange and correlation terms at a slightly lower value of r_s, but still very close to 6.

Figure 2 gives the nonlinear exchange and correlation terms:

$$0.175\, r_s \quad \text{Curve 1}$$

$$-0.0055\, c' r_s^2 \quad \text{Curve 2}$$

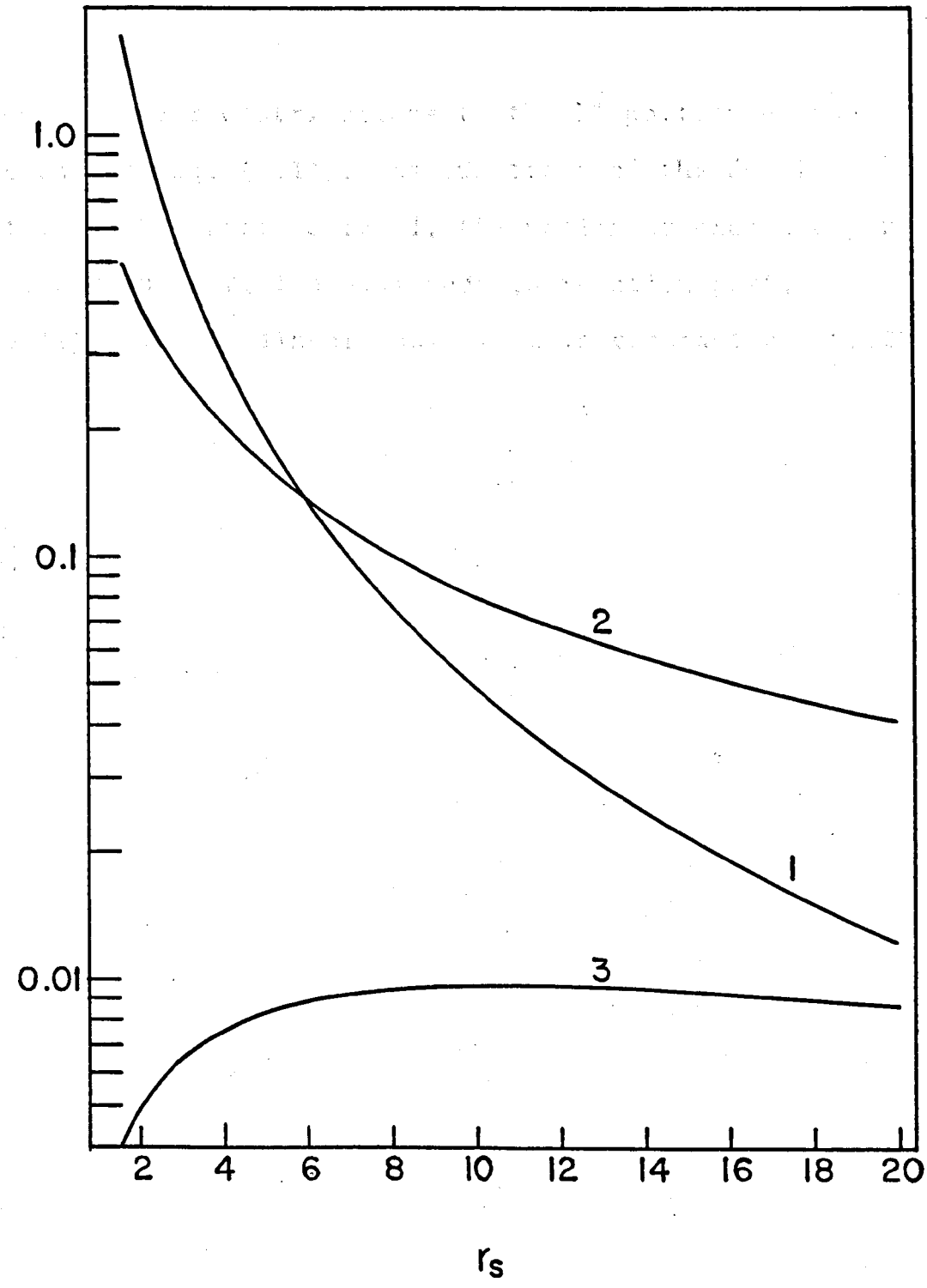
Both contributions ultimately vary as r_s at large r_s, with the correlation term remaining below the exchange term. The nonlinear Pauli term was not plotted because it is constant at -0.529.

We therefore conclude that the collective description of the electron gas is consistent over a density range varying from the usual dense electron gas down to densities at

which the gas crystallizes. The crystallization appears to be the result of the nonlinear corrections (the quadratic terms are not sufficient to produce the lattice), with the exchange term playing the dominant role, followed by the correlation term.

Figure 1

The portions of the k^2 part of the plasmon energy, Eq. (3.14), which do not come from the nonlinear corrections. Curve 1 is the Pauli term, $4.88/r_s^2$, curve 2 is the exchange contributionk $0.815/r_s$, and curve 3 is the correlation term, $0.0251/(c^3-3c^2+2c)$. These are plotted as functions of the density parameter r_s.


1.0
0.1
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2
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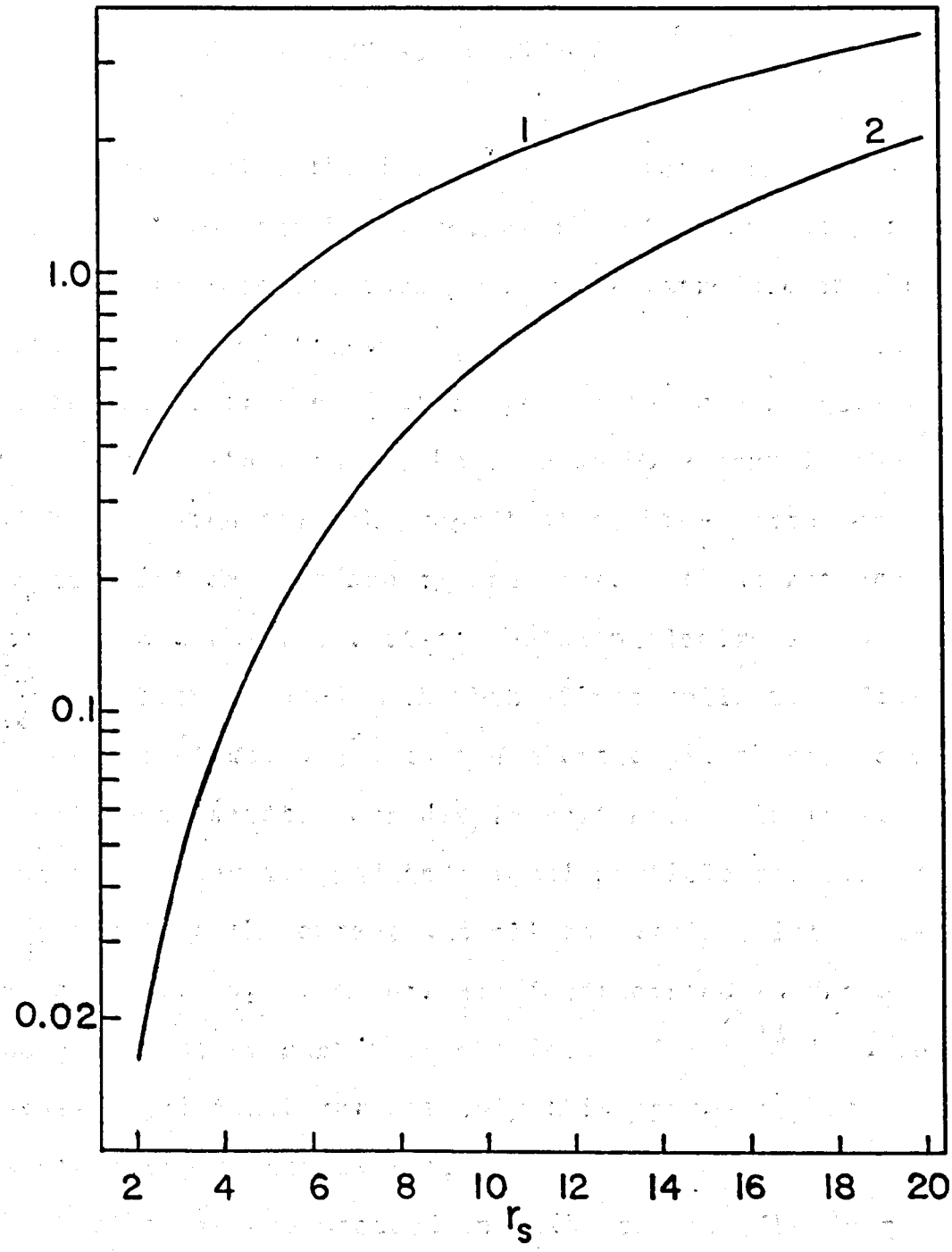
4
6
8
10
12
14
16
18
20
r_s

Figure 2

The nonlinear contributions to the k^2 portion of the plasmon energy, Eq. (3.14). As functions of the density parameter r_s they are: curve 1, the nonlinear exchange part, $0.175r_s$, and curve 2, the nonlinear correlation part, $-0.0055c'r_s^2$. The nonlinear Pauli term is constant at -0.529.


1
2
1.0
0.1
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4
6
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r_s