Theory of two-charged-component Fermi fluids

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A many-body variational method for the ground-state properties of two-charged-component Fermi fluids has been presented. The unique feature of this work is that the theory has been applied to systems with the mass ratio of the two components varying from $m \sim 1$ (electron-hole liquid in Ge or Si) to m = 1836 (liquid-metallic hydrogen). The ground-state structures obtained for these systems are in very good agreement with other theoretical approaches and experiments. We also discuss the collective modes and the possibility of the existence of acoustic plasmons in these liquids.

I. INTRODUCTION

A system of electrons moving against a rigid uniform background of positive charge (the jellium model) has been a classic model¹ for the study of electron correlations in metals. Several theories exist which study the electron gas at zero temperature. There are perturbative methods² and variational methods, $^{3-5}$ which approximately calculate the correlation energy and other quantities with relative success. There are also very accurate Monte Carlo simulations for the electron-gas correlation energy.⁶ There are, however, no experiments for direct comparison with those theoretical results.

The situation is different in the case where one replaces the uniform positive background of the jellium model by a liquid of positively charged particles-the two-chargedcomponent Fermi fluids. At low temperatures and high carrier concentrations, the electrons and holes in semiconductors condense to form a high-density metallic liquid. The electron-hole liquid $(EHL)^{7-10}$ is a collective state of electrons and holes, which is unique in many respects: It is the most metallic of metals and the most quantum of fluids. There are several review articles dealing with the theoretical^{7,9} and experimental¹⁰ aspects of the EHL.

Another interesting example of the two-chargedcomponent Fermi fluids is liquid-metallic hydrogen (LHM).¹¹⁻¹⁵ It is expected that for pressure of the order of 1-10 Mbar, condensed hydrogen will undergo a transition from an insulating molecular crystal phase to a metallic-liquid phase at zero temperature. Thus we have here a system of two interpenetrating Fermi fluids of electrons and protons, which differs from the EHL $(m \sim 1)$ with its vastly different masses between the two species $(m_p/m_e = 1836.109).$

In this paper, we will present a variational calculation of the ground-state properties of these two systems. The formalism to be presented below stem from a generalization^{16,17} of the many-body variational approach for the single-component case¹⁸⁻²⁰ of bosons. In the case of fermions, we select a variational wave function of the Jastrow form

$$\Psi = F_{11}F_{22}F_{12}\Phi_1\Phi_2 , \qquad (1)$$

where $F_{\alpha\beta}$ are symmetric correlation operators and of the form

$$F_{\alpha\beta} = \prod_{i,j} f_{\alpha\beta}(|\vec{\mathbf{r}}_{\alpha,i} - \vec{\mathbf{r}}_{\beta,j}|) , \qquad (2)$$

and Φ_1 (Φ_2) is a determinant of single-particle momentum-spin states for electrons (holes). For bosons, Φ_1 and Φ_2 are set to unity and we have a very reliable method, the hypernetted-chain (HNC) diagrammatic expansion, at our disposal. $^{16-20}$ In the case of fermions, the generalization of the Fermi-HNC scheme of Fantoni and Rosati²¹ is very complicated for computational purposes. Instead, one can adopt a simpler approach by $Lado^{22-24}$ which has been found to be quite accurate for the electron gas^{3,5} and for liquid helium.²⁴

In Sec. II we briefly introduce the basic formalism needed to study the ground-state properties of the boson or fermion mixtures. In Sec. III the Euler-Lagrange equations are solved for different values of the masses of the two species. The corresponding results for EHL's and the LMH are discussed and compared with the other theoretical approaches and experimental results. Section IV addresses the collective modes of the systems studied. The plasmon mode and the most interesting "acoustic-plasmon mode" are discussed. We briefly give our conclusions in Sec. V. Some of the results for EHL's and LMH have been reported earlier.25,26

II. BASIC FORMALISM

For a system of two interpenetrating charged fluids of electrons and holes, the overall change neutrality requires that $e_1N_1 + e_2N_2 = 0$, where e_α and N_α are the charge and particle number of the species α . In the present case, $e_1 = -e_2 = e$, the electron charge; therefore, the partial densities of the two species have to be the same. The density of electrons (or holes) per unit volume is $\rho = k_F^3 / 3\pi^2$ where k_F is the radius of the Fermi sphere. In terms of the dimensionless parameter r_s , the electron (or hole) density is expressed as

 $\rho = \left[\frac{4\pi}{3}a_0^3r_s^3\right]^{-1}.$

The correlation functions (2) are written explicitly as

$$F_{\alpha\beta} = \exp \frac{1}{2} \sum_{\substack{i,j=1\\i < j}}^{N} u_{\alpha\beta} (\mid \vec{\mathbf{r}}_{\alpha,i} - \vec{\mathbf{r}}_{\beta,j} \mid) , \qquad (3)$$

 $N_1 = N_2 = N$ being the number of electrons or holes, and the HNC equations relate $u_{\alpha\beta}(r)$ and the partial-pair correlation functions $g_{\alpha\beta}(r)$ and the static structure functions $S_{\alpha\beta}(k)$ as^{16,17}

$$u_{\alpha\beta}(r) = \ln[g_{\alpha\beta}(r)] - [g_{\alpha\beta}(r) - 1] + C_{\alpha\beta}(r)$$
(4)

and

$$S_{\alpha\beta}(k) - \delta_{\alpha\beta} = C_{\alpha\beta}(k) + \sum_{\gamma=1,2} \left[S_{\alpha\gamma}(k) - \delta_{\alpha\gamma} \right] C_{\beta\gamma}(k) .$$
 (5)

We now introduce the Lado approach^{3,22,23} by writing the spin-averaged ideal-gas probability density as

$$|\Phi_{\alpha}|^{2} = \exp \sum_{\substack{i,j=1\\i < j}}^{N} u_{\alpha\alpha}^{0}(|\vec{\mathbf{r}}_{\alpha,i} - \vec{\mathbf{r}}_{\beta,j}|).$$
(6)

The major advantage of this approximation is that one can again use the HNC equations (4) and (5) to evaluate the radial distribution functions. The most common way to obtain $u_{\alpha\alpha}^{0}(r)$ is to start with the well-known result for the nondynamical correlations,

$$g_F(r) = 1 - \frac{1}{2} [3(\sin x - x \cos x)/x^3]^2, \quad x = k_F r$$
(7)

and the ideal-gas structure factor,

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$$S_{F}(k) = \begin{cases} \frac{3}{4} (k/k_{F}) - \frac{1}{16} (k/k_{F})^{3}, & k \le 2k_{F} \\ 1, & k > 2k_{F} \end{cases}$$
(8)

and invert the HNC equation, using the exact result for $g_F(r)$ to obtain $u_{\alpha\alpha}^0(r)$.

In the Lado-HNC approach the energy per electron is now given by

$$E = \frac{3}{5} \frac{\hbar^2 k_F^2}{2m_1} (1+M) + \frac{\hbar^2 \rho}{8m_1} \left[\int g_{11}^{-1} (\vec{\nabla}g_{11})^2 d\vec{r} + M \int g_{22}^{-1} (\vec{\nabla}g_{22})^2 d\vec{r} + (1+M) \int g_{12}^{-1} (\vec{\nabla}g_{12})^2 d\vec{r} \right] - \frac{\hbar^2}{8m_1} \frac{1}{(2\pi)^3 \rho} \int [S_{11}^2 + MS_{22}^2 + (1+M)S_{12}^2 - (MS_{11} + S_{22})/D - 3S_{11} - 3M(S_{22} - 1) + 3]k^2 d\vec{k} + \frac{\hbar^2 \rho}{8m_1} \int \{ [2\nabla^2 u_{11}^0(r) + (\vec{\nabla}u_{11}^0)^2]g_{11}(r) + M[2\nabla^2 u_{22}^0(r) + (\vec{\nabla}u_{22}^0)^2]g_{22}(r) \} d\vec{r} + \frac{1}{2}\rho e^2 \int (g_{11} + g_{22} - 2g_{12})r^{-1} d\vec{r} ,$$
(9)

 $M = m_1/m_2$, and $D(k) = S_{11}S_{22} - S_{12}^2$. The next step is to find the condition for (9) to be minimum with respect to arbitrary variations of $g_{\alpha\beta}(r)$. This is written in the form

$$[-\nabla^2 + v_{\alpha\beta}(r) + W_{\alpha\beta}(r)]g_{\alpha\beta}^{1/2}(r) = 0, \qquad (10)$$

where the bare interactions $v_{\alpha\beta}(r)$ and the induced potentials $W_{\alpha\beta}(k)$ are given in Refs. 25 and 26. Once the optimum $g_{\alpha\beta}(r)$ are obtained by solving (10), the pressure can readily be obtained from $^{16-18,20}$

$$\frac{P}{\rho} = E - (1+M)\frac{\hbar^2 k_F^2}{5m_1} - \frac{\hbar^2}{8m_1} \frac{1}{(2\pi)^3 \rho} \int \left[\frac{1+M}{D} + \frac{3(S_{22}+MS_{11})}{D} + (S_{11}+MS_{22}) + \frac{(1+M)S_{11}S_{22}}{D^2} - \frac{S_{22}+MS_{11}}{D^2} - 3(1+M) \right] k^2 d\vec{k} .$$
(11)

The expression given above depends only on the structure functions which are obtained from the solutions $g_{\alpha\beta}(r)$ of the Euler-Lagrange (EL) equation (10) and are therefore numerically very convenient. In the case of the boson fluid, this expression has been shown to be equivalent to the pressure obtained from the virial theorem.^{18,20}

The energy expression (9) contains only the two-body distribution functions and can be improved by including the three-body distribution functions.^{3,5,24} However, this improvement in the energy values does not have any noticeable effect on the ground-state structures of the system. One could, therefore, calculate the distribution func-

tions in the present scheme and include the effects of three-body distribution functions from, e.g., Ref. 3. This step has indeed been followed by us in our published results earlier.^{25,26} However, at very low densities, e.g., densities corresponding to the isotropic EHL, the contributions from the three-body distributions were found to be insignificant.²⁷ The energy values to be presented below are obtained from (9).

III. GROUND-STATE PROPERTIES

The ground-state properties of the two-chargedcomponent Fermi fluids are now obtained by solving the

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EL equation (10) and the energy equation (9). The method of solving the EL equations is to generalize the linearization method used earlier for the single-species systems.²⁰ In the following, we will present the numerical results for the two interesting systems which cover the hole-to-electron mass range $m \sim 1-2000$, viz., the EHL and LMH.

A. Electron-hole liquid

We begin our study of the ground-state properties of the EHL by considering the case of isotropic nondegenerate energy bands for the electrons and holes.^{7,8} The most convenient unit for the energy in this case is the binding energy E_x of an exciton—a single bound electron-hole pair. This is given by the standard hydrogenic formula $E_x = e^2/2a_x$, where $a_x = \hbar^2/\mu e^2$ is the Bohr radius of an exciton and μ is the reduced mass, $\mu^{-1} = m_e^{-1} + m_h^{-1}$. The Hartree-Fock energy in this case can readily be obtained as (kinetic plus exchange)

$$E_{\rm HF} = E_k + E_{\rm ex}$$
,

where

$$E_{k} = \frac{3}{5} (E_{F}^{e} + E_{F}^{h}) = 2.21/r_{s}^{2} ,$$

$$E_{ex} = -3e^{2}k_{F}/2\pi = -1.382/r_{s}^{2} .$$
(12)

Here $E_F^{e,h}$ is the Fermi energy of the electrons (holes).

In Fig. 1, we have plotted the solutions $g_{\alpha\beta}(r)$ of (10) for various values of the electron-hole mass ratio. As the hole mass increases a noticeable amount of short-range ordering takes place in the hole-hole correlation functions $g_{h-h}(r)$. Therefore, with the increase in hole mass, the system changes continuously to a state with ordered heavy particles.²⁵ The maximum of the *e*-*h* correlation functions $g_{e-h}(0)$, the enhancement factor, is just the ratio of the



FIG. 1. Pair correlation functions $g_{e-e}(r)$, $g_{h-h}(r)$, and $g_{e-h}(r)$ for the EHL vs $k_F r$. Results are given for different values of m_h/m_e at $r_s = 1$. Note the shift of the origin along $k_F r$ for different m.



FIG. 2. Enhancement factor $g_{e,h}(0)$ as a function of r_s for the EHL in stressed Ge. Solid circles are the experimental results from Ref. 28. CP: present work and Ref. 25; VBS: Ref. 8; HA and RPA: Ref. 7.

electron density on the hole to the mean density. This quantity has recently been measured²⁸ for the EHL in Ge under uniaxial stress. As is known, this system, in the high-stress limit, closely resembles the isotropic case we have been discussing.⁷ Therefore, as stressed in Ref. 28, this result provides a sensitive and valuable test for the



FIG. 3. Ground-state energy and pressure vs r_s for the EHL.

predictions of many-body approximation schemes. In Fig. 2, we have compared our variational results with the experimental results and other theoretical results. In the figure, HA and RPA refer to the standard Hubbard and the random-phase approximations,^{7,8} and VBS refers to a self-consistent scheme by Vashishta, Bhattacharyya, and Singwi.⁸ The comparison is strong evidence that our variational result [note CP, for Chakraborty and Pietiläinen (Ref. 25), present in Fig. 2] quite accurately predicts the structure of the EHL for the density range of interest.

Recently, there have been some experimental studies of the decay kinetics of electron-hole drops.²⁹ These studies provide information about the intrinsic recombination of the electron-hole drops. Considering the case of Si under uniaxial stress, and assuming that the most probable decay mechanism in this case is Auger recombination, one obtains the density variation of $g_{\alpha\beta}(0)$. Here too, our variational result agrees very well³⁰ with the experimental results of Ref. 29.

The ground-state energy and pressure (in units of exciton Rydbergs) are plotted in Fig. 3 as a function of r_s for different values of the hole-to-electron mass ratio. The case of $m_e/m_h = 1$ has been compared with two other theoretical approaches: Brinkman, Rice, Anderson, and Chui (BRAC) from Ref. 7 and VBS from Ref. 8. All the calculations predict a local energy minimum at an equilibrium density; the minimum energy in all calculations, however, lies above the energy of free excitons (-1 Ry in)the isotropic case). Therefore, the metallic state can be metastable minimum, but not the ground state. This conclusion remains unaltered even for $m_h >> m_e$, as our results indicate. In fact, it is known from the study of an-isotropic cases⁷⁻⁹ that the effects of band anisotropy, coupling between degenerate valence bands, and other effects tend to stabilize the metallic liquid in the semiconductors. A study of these effects is however, beyond the scope of the present work. For a detailed report on these effects in EHL's for various semiconductors, we refer to the work of Rice.7,31

B. Liquid-metallic hydrogen

Unlike EHL, LMH has yet to be realized in the laboratory. However, there are several theoretical calculations which raise the possibility of a stable (or metastable) phase of LMH at low temperatures and at least for a certain narrow density range. Recently, Mon et al.¹² calculated the upper bounds for the ground-state energies of liquid and solid phases using the Jastrow-Slater many-body variational ansatz and Monte Carlo techniques. The calculations were based on a model of pair interactions between protons, and therefore take into account only the secondorder effects in the electron-proton screened interaction. They concluded that, while at the densities corresponding to $r_s = 0.8 - 1.6$, the solid phase was preferred, at $r_s \approx 1.6$, the liquid phase could not be ruled out. Later, Chakravarty and Ashcroft¹³ calculated a variational upper bound to the ground-state energies of the LMH and included the important third-order effects in the electron-proton screened interaction. They also concluded that the possibility of a liquid metallic phase cannot be ruled out at



FIG. 4. Pair correlation functions $g_{e-e}(r)$, $g_{p-p}(r)$, and $g_{e-p}(r)$ vs $k_F r$ for LMH. Dashed lines for $g_{p-p}(r)$ are by Chakravarty and Ashcroft (Ref. 13).

T=0 around the density corresponding to $r_s \approx 1.64$. There is also a recent work¹⁵ where it has been argued that, because of the differing roles of electronic screening in solid and fluid states, metallic hydrogen will remain a fluid at all pressures.

In our calculations of the liquid phase, we have solved again the EL equation (10) for the electron-proton mass ratio and Coulomb interaction for the electron-electron, proton-proton, and electron-proton pairs. In Fig. 4, we have plotted the distribution functions $g_{e-e}(r)$, $g_{p-p}(r)$, and $g_{e-p}(r)$ as functions of $k_F r$ for different values of r_s . Comparing these results with those of Fig. 1, we find that the general behavior of the distribution functions for the LMH is apparently a continuous development of the EHL case as the hole mass increases. The proton-proton distribution functions are compared with the results of Chakravarty and Ashcroft¹³ for $r_s = 0.8$ and 1.36 and good agreement is obtained.

Finally, we present in Fig. 5 the energy and pressure



FIG. 5. Ground-state energy and pressure vs r_s for LMH. Dashed line is by Chakravarty and Ashcroft (Ref. 13).

values for the LMH as a function of r_s using (9) and (11) and the optimum solutions $g_{\alpha\beta}(r)$. We also present the energy values obtained by Chakravarty and Ashcroft. As we mentioned above, a more improved result has been given in Ref. 26; however, the difference between the present result and that of Ref. 13 is quite small around the density $r_s = 1.6$ and decreases with r_s . We have not been able to obtain any stable solutions for $r_s > 1.6$, where the pressure is expected to vanish. This fact might be related to the rapid enhancement of the compressibility in that density range,¹⁵ leading to difficult numerical problems encountered earlier.²⁰ There are other instabilities³² which also have to be studied at these densities.

IV. COLLECTIVE MODES

We now consider the collective excitation modes in the two-charged-component Fermi fluids. First we have the usual plasmon mode where the particles oscillate out of phase. The uniform displacement of the two species toward the opposite direction from each other gives rise to the depolarizing field. The strength of this field acts as the restoring force which produces the plasma oscillation¹ with a nonzero frequency even at k=0. In a two-component fluid, this frequency is

$$\omega^2 = \omega_{p_1}^2 + \omega_{p_2}^2 , \qquad (13)$$

where $\omega_{p_i}^2 = 4\pi\rho e_i^2/m_i$ is the plasma frequency of the *i*th species. In this case, the charge densities of the two species add together, resulting in a greater restoring force and higher frequency compared to either of the separate plasmas.

The most interesting mode in a two-charged-component



FIG. 6. Excitation spectrum for the EHL. Dashed-dotted lines represent the maximum energy of the particle-hole pair excitations as explained in the text.



FIG. 7. Same as in Fig. 6, but for LMH at different values of r_s .

Fermi fluid is, however, the acoustic-plasmon mode,^{33,34} where the particles oscillate in phase with the light particles following the motion of the heavy particles in such a way as to screen out the field of heavy particles. However, for the Landau damping of the acoustic plasma wave (by the faster species) to be relatively small, one should have $m_1/m_2 \ll 1$, which means the Fermi velocities of the two species will have to be quite different from one another. In fact, this is a necessary condition, as earlier studies indicate,^{33,34} for the existence of a well-defined acoustic wave traveling at a speed that is essentially the geometric mean of the two Fermi velocities.

Recently, Vignale and Singwi³⁵ studied the collective modes in the isotropic EHL for different values of hole-to-electron mass ratio, $m = m_h/m_e$. They obtained a minimum value of m to be $7 < m_0 < 10$, below which the acoustic mode does not exist.

Oliva and Ashcroft¹⁴ studied the collective modes in the LMH in a generalized Landau-Silin-Boltzmann—equation approach. They noticed that the acoustic-plasmon mode will be Landau-damped due to (at least) electron particle-hole pair excitations. However, on account of the large component mass ratio, it would be difficult for the light electrons to retard the heavy, slowly oscillating protons. Therefore, the Landau damping of LMH by the electrons is expected to be relatively small.

In order to study the collective modes in a twocharged-component Fermi fluid with variable mass ratio, we adopt the generalization of the Bijl-Feynman equation for the two-component system.³⁶ Then the two branches of the excitation spectrum are given as the eigenvalues of the matrix $S^{-1}(k)\epsilon_0(k)$, where S is the matrix with component $S_{\alpha\beta}(k)$ and $\epsilon_0(k) = \text{diag}(\hbar^2 k^2/2m_{\alpha})$. The eigenvalues are readily obtained as^{26,36}

$$\epsilon_{1,2}/\epsilon_F = (q_0^2/2D) \{ (\sigma S_{11} + S_{22}) \\ \pm [(\sigma S_{11} + S_{22})^2 - 4\sigma D]^{1/2} \}, \quad (14)$$

where $\sigma = m_1/m_2$, $q_0 = k/k_F$, and ϵ_F is the electron Fermi energy.

In Fig. 6 we have plotted the two branches of the excitation spectrum ϵ_1 and ϵ_2 in units of electron Fermi energy, as a function of q_0 for m = 1, 2, 10, 50, and the proton-to-electron mass ratio. The dashed-dotted lines are the threshold energy for the onset of Landau damping due to excitation of particle-hole pairs of type 1 $[\omega_1/\epsilon_F]$ $=q_0(q_0+2)$] and that of type 2 $[\omega_2/\epsilon_F=q_0(q_0+2)\sigma]$ (the curve in the upper half of the figure and the curves in the lower half of the figure, respectively). The acoustic mode does not seem to exist for m < 10. The cutoff value q_c of k, where the acoustic mode enters the particle-hole continuum of the heavier species, increases with the mass ratio, while it slowly decreases for the plasmon mode as it enters the particle-hole continuum of the lighter species. Thus a degenerate two-charged-component Fermi fluid might support an acoustic plasma wave if one of the species is quite heavier than the other species. In Fig. 7 we have plotted the collective modes in LHM for different values of r_s . The value of q_c for the acoustic mode is apparently independent of density, but increases for the plasmon mode. While the general shape of the curves for the acoustic mode remains the same for different values of r_s , the small-k behavior changes quite rapidly with r_s . In fact, for $r_s \ge 1.6$, the acoustic mode remains below the maximum energy of the particle-hole pair excitations (dashed-dotted line) for 0 < q < 0.5. Therefore, at these densities the low-k acoustic plasmons possibly decay into single-particle excitations. It should be recalled that at these densities we have not been able to obtain any stable solutions of Eq. (10).

The collective modes in LMH have also been studied within the "mean-spherical-approximation" scheme³⁷ using the Bijl-Feynman equations. While this is a high-density-limit approach, it leads to a qualitatively correct picture for the collective modes at the densities of interest. The method is also convenient for a formal study of the collective modes.

Finally, it would be interesting to study the collective excitations of the two-charged-component fluids when the component species are bosons. In Ref. 17 we have studied the ground-state structures of such a system. The structure functions obtained in that paper are given in Fig. 8 for $r_s = 0.126$ and 1.26. (In Ref. 17, r_s corresponds to the total density of the system, and hence $r_s = 0.1$ and 1.0, respectively.) We have calculated the two branches of the excitation spectrum from Eq. (14). As shown in Fig. 9 one obtains the usual plasma-type excitation, while the other mode is free-particle type. This is exactly what was expected³⁸ in these systems.

V. CONCLUSIONS

Using a many-body variational approach for twocharged-component Fermi fluids, we have been able to study the various ground-state properties of two very in-



FIG. 8. Partial structure functions $S_{\alpha\beta}(q)$ vs $q = ka_0r_s$ for the two-charged-component Bose fluids (from Ref. 17).

teresting systems: EHL's and LMH. Some of the results obtained here are compared with recent experimental and theoretical results and are found to be in good agreement with them. The study of elementary excitations in these systems reveals the possibility that the system might support an acoustic plasma wave if one of the species is much heavier than the other. Finally, the present work can be extended to two dimensions for these systems, where the short-range correlations play a very important role. Such calculations for the layered EHL will be published elsewhere.³⁹



FIG. 9. Excitation spectrum for the two-charged-component Bose fluids.

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