

Two-component Fermi-liquid theory: Equilibrium properties of liquid metallic hydrogen

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(Received 1 August 1980)

Recent studies point to the possibility that condensed hydrogen will undergo a transition from an insulating molecular crystal phase to a metallic *liquid* phase at zero temperature and high pressure. Liquid metallic hydrogen (LMH) comprising interpenetrating proton and electron fluids, would constitute a two-component Fermi liquid having both long-range, species-dependent bare interactions and a very high component-mass ratio. We examine the low-temperature equilibrium properties of LMH (assuming that it is "normal") by means of a generalization to the case of two components of the phenomenological Landau Fermi-liquid theory. The general two-component formalism is discussed in some detail. Estimates for the relevant phenomenological Landau parameters for LMH are made, and results for low-temperature specific heat, compressibility, thermal expansion coefficient, and spin susceptibility are given. The specific heat and thermal expansion coefficient are found to be vastly greater in the liquid than in the corresponding solid due to the presence of proton quasiparticle excitations in the liquid. The possibility of a *negative* expansion coefficient of the liquid cannot be dismissed.

I. INTRODUCTION

It has been realized for some time that any ordinary macroscopic substance subjected to sufficiently high pressure can exist in a metallic state. Historically, one of the most tantalizing prospects for metallization has been the element hydrogen.¹ At atmospheric pressure and at temperatures of the order of 1 K, hydrogen exists in an insulating molecular solid phase.² At these temperatures it is believed that the transition to the atomic metallic phase occurs at a pressure in the range $\sim 1-10$ Mbar.¹

In general, it has been assumed that through the low-temperature insulator-to-metal transition, hydrogen remains a solid. However, the possibility of transforming from the solid insulating phase into a *liquid* metallic phase has been put forward by Brovman *et al.*³ on the basis of an exhaustive study of the relative energies of possible structural modifications of H at various densities.⁴ They included electronic screening in the Hubbard approximation and third-order effects in the electron-proton screened interaction. Hammerberg⁵ subsequently calculated the ground-state energies of the possible liquid and several simple cubic solids within perturbation theory using the Geldart-Vosko modification of Hubbard screening, and also by taking into account second- (fourth-) order effects in the electron-proton screened interaction in the liquid (solid). He concluded that the ground-state energies of liquid and solid phases were indistinguishable within the errors of the approximations for certain densities. More recently, Mon *et al.*⁶ calculated upper bounds for ground-state energies of liquid and solid phases with a Jastrow-function variational ansatz and Monte Carlo techniques, and taking

into account second-order effects in the electron-proton screened interaction. They concluded that at a density corresponding⁷ to $r_s \approx 1.6$ the liquid phase could not be ruled out, though at higher densities, $r_s = 0.8-1.6$, the solid phase was preferred. Most recently, Chakravarty and Ashcroft⁸ calculated a variational upper bound to the ground-state energies of metallic hydrogen including important third-order effects in the electron-proton screened interaction and again concluded that the possibility of a $T=0$ liquid metallic phase cannot be ruled out for $r_s \approx 1.64$.

Naturally, one might consider undertaking even more elaborate calculations to shed further light on the question of the preferred phase.⁹ In due course the issue may be resolved in the laboratory, with efforts toward attaining \sim Mbar pressures already actively underway.¹⁰ But faced as we are with significant preliminary evidence pointing to the possibility of a stable (or perhaps metastable) phase of liquid metallic hydrogen at low temperatures and at least for a certain narrow density range, it seems appropriate to consider the behavior such a substance might exhibit. For instance, will liquid metallic hydrogen display any striking or novel behavior as compared with other quantum liquids such as ³He, ⁴He, electrons in a metal, etc.? Or, in a more utilitarian vein, we may ask how the behavior of the putative liquid metallic phase compares with that of the corresponding solid metallic phase. Any differences in behavior (e.g., in resistivity versus temperature) will be potentially useful in distinguishing experimentally between the two phases when metallization is finally achieved.¹¹

Liquid metallic hydrogen (LMH), comprising interpenetrating electron and proton fluids, would constitute a two-component Fermi liquid with the

components each charged and each of spin $\frac{1}{2}$.¹² In contrast to the situation prevailing in two other two-component Fermi liquids, namely, nuclear matter (neutrons and protons) and the electron-hole liquid of semiconductors (electrons and holes), in LMH the masses of the two species are vastly different. In further contrast to nuclear matter, the bare interparticle interactions in LMH are long range and species dependent. Long-range forces will, as usual, require special treatment and will qualitatively affect certain properties as compared to the case with short-range forces. In further contrast to the electron-hole plasma of semiconductors there are no anisotropy (e.g., band-structure-related) effects in LMH. Thus the high density liquid metallic phase of hydrogen appears to correspond to an as yet relatively uninvestigated class of two-component interacting Fermi fluids.

At this stage it should be pointed out that in assuming the existence of a liquid phase, the very interesting question still remains of whether or not it exhibits some form of magnetic, momental, or even spatial (e.g., liquid crystal¹³) ordering. It was pointed out by Ashcroft¹⁴ that a BCS-type argument leads to a relatively high superconducting transition temperature T_c for solid hydrogen. The origin of this can be traced to a high Debye frequency (i.e., low ionic mass) and high electron-phonon coupling. It was subsequently realized¹⁵ that a similar argument could be applied to a liquid phase as well, the point being that the BCS argument (in its simplest form) essentially presupposes only a uniform elastic medium within which long-lived phonons can exist for a wide range of wave vectors. The "long-lived" requirement breaks down in the liquid for smaller wave vectors than in the solid, but the line of reasoning is nevertheless suggestive. Recently, a more sophisticated strong-coupling calculation by Jaffe and Ashcroft¹⁶ confirms the promise of a high T_c in the liquid, though it is as usual difficult to assess the accuracy of such an "ab initio" calculation at present. Further speculation might turn to possible low-temperature superfluid behavior of the "screened" protons or ferromagnetic ordering among the protons.^{8,15}

We do not attempt at this time to resolve the important questions of the existence or properties of possible "ordered" liquid metallic phases of hydrogen. Rather, we address in this paper the somewhat parallel issue of the general low-temperature equilibrium properties of a "normal" (i.e., nonordered) liquid phase of metallic hydrogen. To this end we utilize a generalization of the phenomenological Landau-Fermi-liquid

theory to the case of two very different components. The basic formalism is discussed in Sec. II, and general equilibrium results are given in Sec. III. In Sec. IV estimates of the relevant "Landau parameters" for LMH are given, and these are applied to the general equilibrium results. Equilibrium behavior of solid and liquid phases is then contrasted. We conclude in Sec. V with an overview and outlook for further effort.

II. THE TWO-COMPONENT FERMI LIQUID

We generalize the phenomenological Landau theory of Fermi liquids¹⁷ to the case of a uniform system of two spin- $\frac{1}{2}$ fermion components (labeled 1 and 2).¹⁷ The masses m_1 and m_2 of the two components are, in general, different; the bare interactions are taken to be long-range Coulomb with the charges e_i of the species satisfying $e_1 = -e_2$.¹⁸ Requiring overall charge neutrality, the average number densities n_i of the two species are then equal. This in turn implies that for the corresponding noninteracting system, the Fermi momenta p_{F_i} of the two species (i.e., the momenta of the most energetic particle of each species at temperature $T=0$) are equal: $p_F \equiv p_{F_i} = (3\pi^2 n_i)^{1/3}$. However, the Fermi energies $E_{F_i} = p_{F_i}^2/2m_i$ and Fermi temperatures $T_{F_i} \equiv E_{F_i}/k_B$, (with k_B Boltzmann's constant) will be different on account of the difference in mass. For the case of the noninteracting system corresponding to LMH with the masses differing by a factor of $\sim 10^3$ and at a typical density corresponding to Wigner-Seitz radius of 1.6, the electron and proton Fermi temperatures are $\sim 10^5$ and $\sim 10^2$ K, respectively. One thus expects characteristic degenerate Fermi-gas behavior of both components to be evident at $T \sim 10$ K. When the interactions are restored we would expect in analogy with liquid ^3He (Ref. 17) a lowering by a factor of $\sim 10-10^2$ of the maximum temperature at which degenerate Fermi-liquid behavior will be apparent.

Following the usual assumptions of Landau-Fermi-liquid theory, the entropy of the interacting system at low temperature S is given as a functional of the quasiparticle distribution functions $n_{i\vec{p}\sigma}$, which is of the same form as that for the entropy of a noninteracting system expressed in terms of the bare particle distribution functions. Thus,

$$\begin{aligned}
 S &= S[n_{1\vec{p}\sigma}, n_{2\vec{p}\sigma}] \\
 &= -k_B \sum_{i\vec{p}\sigma} [n_{i\vec{p}\sigma} \ln n_{i\vec{p}\sigma} + (1 - n_{i\vec{p}\sigma}) \ln(1 - n_{i\vec{p}\sigma})],
 \end{aligned}
 \tag{1}$$

where the $n_{i\vec{p}\sigma}$ give the distribution of quasiparticles (qp) of type i , momentum \vec{p} , and z component of spin $\frac{1}{2}$ ($\sigma = \pm 1$).

The equilibrium qp distribution functions $n_{i\vec{p}\sigma}^0$ are determined as those maximizing the entropy subject to the constraint of fixed total particle number of each species $N_i = \sum_{\vec{p}\sigma} n_{i\vec{p}\sigma}$, and fixed total energy $E = E[n_{1\vec{p}\sigma}, n_{2\vec{p}\sigma}]$, in general, an unknown functional to be treated phenomenologically:

$$n_{i\vec{p}\sigma}^0 = \frac{1}{\exp[\beta(\epsilon_{i\vec{p}\sigma} - \mu_i)] + 1}, \quad i=1,2 \quad (2)$$

where $\beta = 1/k_B T$, μ_i is the chemical potential of the i th species in the interacting system, and

$$\epsilon_{i\vec{p}\sigma} = \frac{\delta E}{\delta n_{i\vec{p}\sigma}^0} = \epsilon_{i\vec{p}\sigma}[n_{1\vec{p}\sigma}^0, n_{2\vec{p}\sigma}^0], \quad i=1,2. \quad (3)$$

The $\epsilon_{i\vec{p}\sigma}$ as well as the μ_i are each functionals of both equilibrium qp distribution functions and as such Eq. (2) represents a set of two coupled implicit functional equations for $n_{1\vec{p}\sigma}^0$ and $n_{2\vec{p}\sigma}^0$. The quasiparticle energies for each species thus incorporate effects due to the interactions with particles of the other species as well as with the same. We assume the $\epsilon_{i\vec{p}\sigma}$ are spin independent and smooth as functions of \vec{p} .¹⁹

For the low-temperature equilibrium properties we need to consider the four second variational derivatives of the total energy with respect to the distribution functions:

$$f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{ij} = \frac{\delta^2 E}{\delta n_{i\vec{p}\sigma}^0 \delta n_{j\vec{p}\sigma'}^0} = f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{ij}[n_{1\vec{p}\sigma}^0, n_{2\vec{p}\sigma'}^0], \quad i,j=1,2. \quad (4)$$

f^{ij} may be interpreted as the interaction energy between qp of types i and j . We assume the f^{ij} 's are smooth as functions of \vec{p} and \vec{p}' . Note, in general, $f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{12} \neq f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{21}$, though $f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{12} = f_{\vec{p}'\vec{\sigma}'\vec{\sigma}}^{21}$.²⁰

Since the system is invariant under time reversal (no magnetic field present), $f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{ij} = f_{-\vec{p},-\sigma,-\vec{p}',-\sigma'}^{ij}$. Furthermore, the Fermi surfaces are spherical and are invariant under reflection $\vec{p} \rightarrow -\vec{p}$. Thus

$$f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{ij} = f_{\vec{p}-\sigma,\vec{p}'-\sigma'}^{ij}, \quad i,j=1,2. \quad (5)$$

We then define in the usual way the two independent components of f^{ij} , namely, the spin-symmetric and spin-antisymmetric parts, ${}^s f^{ij}$ and ${}^a f^{ij}$:

$${}^s(a) f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{ij} \equiv \frac{1}{2} (f_{\vec{p}\vec{\sigma}\vec{p}'\vec{\sigma}'}^{ij} + f_{\vec{p}\vec{\sigma}\vec{p}'\vec{\sigma}}^{ij} + f_{\vec{p}\vec{\sigma}\vec{p}'\vec{\sigma}'}^{ij} - f_{\vec{p}\vec{\sigma}\vec{p}'\vec{\sigma}}^{ij}), \quad i,j=1,2. \quad (6)$$

Since the f^{ij} 's incorporate the (spin-independent) equilibrium qp distribution functions, the spin-antisymmetric interspecies f 's must vanish identically:

$${}^a f^{12} = {}^a f^{21} = 0. \quad (7)$$

For low-temperature properties we only need the $\epsilon_{i\vec{p}\sigma}$ and $f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{ij}$ evaluated for $|\vec{p}|, |\vec{p}'| = p_F$. Paralleling the one-component case we introduce the following parametrization. A qp effective mass m_i^* and in turn a qp velocity at the Fermi surface \vec{v}_{F_i} are defined via

$$\vec{v}_{F_i} \equiv \frac{\vec{p}_F}{m_i^*} \equiv \vec{\nabla}_{\vec{p}} \epsilon_{i\vec{p}\sigma} \Big|_{p=p_F}. \quad (8)$$

The qp energies in the vicinity of the Fermi surface are then

$$\epsilon_{i\vec{p}\sigma} \approx \mu_i + \frac{p_F}{m_i^*} (p - p_F) \quad (p \approx p_F). \quad (9)$$

Note that $m_i^* > 0$.²¹

Since the system is isotropic $f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{ij}$ for $|\vec{p}|, |\vec{p}'| = p_F$ can only depend on the angle θ between \vec{p} and \vec{p}' . Then the f^{ij} 's may be expanded in the usual Legendre polynomial series:

$${}^s(a) f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{ij} = \sum_{l=0}^{\infty} {}^s(a) f_l^{ij} P_l(\cos\theta) \quad (|\vec{p}|, |\vec{p}'| = p_F; \quad i,j=1,2). \quad (10)$$

We note that since $f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{12} = f_{\vec{p}\vec{\sigma}\vec{\sigma}'}^{21}$ when $|\vec{p}| = |\vec{p}'|$,

$${}^s(a) f_l^{12} = {}^s(a) f_l^{21}.$$

As in the one-component case, a consequence of Galilean invariance is the existence of a simple relationship between the effective mass and the "f functions".²² Starting with the system at rest and in the equilibrium configuration, we first add a single quasiparticle $\vec{p}\sigma$ of type i at the Fermi surface. By Galilean invariance, the energy change $\epsilon'_{i\vec{p}\sigma}$ as viewed from a frame (primed) moving with velocity \vec{u} is related to the resulting energy change $\epsilon_{i\vec{p}\sigma}$ as viewed from the lab frame (unprimed) by (to order u)

$$\epsilon'_{i\vec{p}\sigma} = \epsilon_{i\vec{p}\sigma} + \frac{m_i - m_i^*}{m_i^*} \vec{p} \cdot \vec{u}, \quad (11)$$

where we have used $\vec{p}' = \vec{p} - m_i \vec{u}$, replaced $\vec{p} - \vec{p} + m_i \vec{u}$, and expanded $\epsilon_{i\vec{p}+m_i\vec{u},\sigma}$ using Eq. (8). But then, to order u ,

$$\begin{aligned} \epsilon'_{i\vec{p}\sigma} &= \epsilon_{i\vec{p}\sigma} [n_{1\vec{p}+m_i\vec{u},\sigma}^0, n_{2\vec{p}+m_i\vec{u},\sigma}^0] \\ &\approx \epsilon_{i\vec{p}\sigma} [n_{1\vec{p}\sigma}^0 + m_i \vec{u} \cdot \vec{\nabla}_{\vec{p}} n_{1\vec{p}\sigma}^0, n_{2\vec{p}\sigma}^0 + m_i \vec{u} \cdot \vec{\nabla}_{\vec{p}} n_{2\vec{p}\sigma}^0] \end{aligned} \quad (12)$$

Expanding the right-hand side of Eq. (12) in a functional Taylor series about $n_{i\vec{p}\sigma}^0$, using Eqs. (2), (4), and (5), then substituting into Eq. (11), we find, equating terms linear in \vec{u} , the desired relation:

$$\frac{m_i^*}{m_i} = \frac{1}{1 - (V p_F^2 / 3\pi^2 \hbar^3) ({}^s f_1^{ii} m_i + {}^s f_1^{ij} m_j)}, \quad j \neq i \quad (13)$$

with V the volume.

The requirement of thermodynamic stability, i.e., that $E - \mu_1 N_1 - \mu_2 N_2$ be a minimum for the equilibrium distribution functions, leads (at $T \approx 0$) to bounds on the possible values of the Landau f parameters.²² The change in $E - \mu_1 N_1 - \mu_2 N_2$ produced by arbitrary distortions $\delta n_{i\vec{p}\sigma}$ is given to second order by [Eqs. (3), and (4)]

$$\begin{aligned} \delta(E - \mu_1 N_1 - \mu_2 N_2) \approx & \sum_{i\vec{p}\sigma} (\epsilon_{i\vec{p}\sigma} - \mu_i) \delta n_{i\vec{p}\sigma} \\ & + \frac{1}{2} \sum_{i\vec{p}\sigma\vec{p}'\sigma'} f_{i\vec{p}\sigma\vec{p}'\sigma'}^{ij} \delta n_{i\vec{p}\sigma} \delta n_{j\vec{p}'\sigma'} \end{aligned} \quad (14)$$

We choose $\delta n_{i\vec{p}\sigma} = n_{i\vec{p}\sigma} - n_{i\vec{p}\sigma}^0$ ($T=0$) such that $n_{i\vec{p}\sigma}$ is of the form $n_{i\vec{p}\sigma} = \Theta(p_{iF}(\theta, \sigma) - p)$ where $p_{iF}(\theta, \sigma)$ is an arbitrary distortion of the Fermi surface. Here $\Theta(x) = 1$ for $x > 0$ and vanishes otherwise, and θ is the polar angle of \vec{p} . We expand $\delta n_{i\vec{p}\sigma}$ in Eq. (14) to second order in $\delta p_{iF} \equiv p_{iF}(\theta, \sigma) - p_F$, and expand

$$\delta p_{iF}(\theta, \sigma) = \frac{m_i^*}{p_F} \sum_l p_{il\sigma} P_l(\cos\theta).$$

In order that the resulting quadratic form in the $p_{il\sigma}$'s be positive definite we finally obtain

$$1 + \frac{\nu_i {}^a f_i^{ii}}{2l+1} > 0, \quad (15a)$$

$$1 + \frac{\nu_i {}^s f_i^{ii}}{2l+1} > 0, \quad i=1,2 \quad (15b)$$

and

$$\left(1 + \frac{\nu_1 {}^s f_1^{11}}{2l+1}\right) \left(1 + \frac{\nu_2 {}^s f_2^{22}}{2l+1}\right) - \frac{\nu_1 \nu_2}{(2l+1)^2} ({}^s f_1^{12})^2 > 0, \quad (15c)$$

where ν_i , the density of states at the Fermi surface is given by

$$\nu_i = \frac{m_i^* p_F V}{\pi^2 \hbar^3}. \quad (16)$$

These inequalities can be viewed as statements that there can be neither too much interspecies repulsion [Eq. (15c)] nor too much inter- or intraspecies attraction [Eqs. (15c) or (15b) and (15a)] in order that the system remain normal. The result of the former is phase separation (when interactions are short range) while the result of the latter includes the possibilities of, for example, formation of atomiclike or more complex "bound" entities (including a crystal) or occurrence of spin or-

dering (see below).

We have so far ignored the implications of the long-range Coulomb nature of the bare interactions in the problem. We consider first the interaction Hamiltonian of a two-component system (having equal numbers of each species), where the bare interactions are Yukawa: $U_{11} = U_{22} = -U_{12} = e^{-\mu r}/r \equiv U_\mu(r)$. The original Coulomb system is recovered in the limit $N_i \rightarrow \infty$, $V^{1/3} \rightarrow \infty$, with $N_i/V = \text{const}$, and $\mu \rightarrow 0$ such that $V^{1/3}\mu \gg 1$. The interaction Hamiltonian \hat{H}_I may be written as

$$\begin{aligned} \hat{H}_I = & \frac{1}{2V} \sum_{\vec{q}} [\hat{\rho}_1(\vec{q}) \hat{\rho}_1(-\vec{q}) - \hat{N}_1 + \hat{\rho}_2(\vec{q}) \hat{\rho}_2(-\vec{q}) - \hat{N}_2 \\ & - 2\hat{\rho}_1(\vec{q}) \hat{\rho}_2(-\vec{q})] U_\mu(q), \end{aligned} \quad (17)$$

where $\hat{\rho}_i(\vec{q})$ is the Fourier transform of the density operator of the i th species, where $U_\mu(q)$ is the Fourier transform of $U_\mu(r)$: $U_\mu(q) = 4\pi/(q^2 + \mu^2)$, and where \hat{N}_i is the number operator of the i th species.

Noting that $\hat{\rho}_i(0) = \hat{N}_i$ and that $N_1 = N_2$ for the state under consideration we see that the $\vec{q}=0$ term in the interaction energy vanishes identically when the above Coulomb limit is taken. Thus we may just as well take for \hat{H}_I for the original Coulomb problem the above expression with the replacement of $U_\mu(q)$ (together with the taking of the Coulomb limit) by $U'(q)$ defined as $1/q^2$ for $q \neq 0$, and 0 for $q=0$.

Actually, we point out that it is formally possible to use the original $U_\mu(q)$ with $\mu=0$ in \hat{H}_I directly. This leads to the f^{ij} 's having a divergent part as well as a well-behaved "short-range" part. However, in any formal expression for a thermodynamic property, the combinations of the f^{ij} 's which appear must be of such a form that these divergent parts cancel identically leaving a physically meaningful result (see the discussion of the compressibility below). Nonetheless, in what follows we generally view the ϵ_i 's and f^{ij} 's as appropriate to the choice $U'(q)$.

III. EQUILIBRIUM PROPERTIES—GENERAL

We consider first the specific heat at constant volume

$$C_V = \frac{T}{V} \frac{\partial S}{\partial T} \Big|_V.$$

This is obtained using the entropy equation (1) evaluated at the equilibrium qp distribution functions equation (2) together with expression (9) for the qp energy near the Fermi surface:

$$C_V = \frac{p_F k_B^2}{3\hbar^3} (m_1^* + m_2^*) T, \quad T \ll \min(T_{F_1}, T_{F_2}) \quad (18)$$

i. e., as expected, the specific heat is linear in temperature with coefficient proportional to the sum of the effective masses.

Next, for a general two-component system, the isothermal compressibility K_T may be written as:

$$K_T \equiv -\frac{1}{V} \frac{\partial V}{\partial p} \Big|_T = \left(n_1^2 \frac{\partial \mu_1}{\partial n_1} \Big|_{n_2, T, V} + n_1 n_2 \frac{\partial \mu_1}{\partial n_2} \Big|_{n_1, T, V} + n_1 n_2 \frac{\partial \mu_2}{\partial n_1} \Big|_{n_2, T, V} + n_2^2 \frac{\partial \mu_2}{\partial n_2} \Big|_{n_1, T, V} \right)^{-1}. \quad (19)$$

To make contact with Landau theory we use Eq. (2) to relate variations in $n_{i\vec{p}\sigma}$ about the equilibrium value to variations in the $\epsilon_{i\vec{p}\sigma}$ and μ_i :

$$\delta n_{i\vec{p}\sigma} = \frac{\partial n_{i\vec{p}\sigma}^0}{\partial \epsilon_{i\vec{p}\sigma}} (\delta \epsilon_{i\vec{p}\sigma} - \delta \mu_i). \quad (20)$$

But the changes $\delta \epsilon_{i\vec{p}\sigma}$ associated with hydrostatically compressing the system are self-consistently related to changes in $\delta n_{i\vec{p}\sigma}$ (in this case spin independent) via [see Eqs. (3), (4), and (6)]

$$\delta \epsilon_{i\vec{p}\sigma} = 2 \sum_{\vec{p}'} (s_{\vec{p}\vec{p}'}^{i1} \delta n_{1\vec{p}'} + s_{\vec{p}\vec{p}'}^{i2} \delta n_{2\vec{p}'}) \quad (21)$$

Noting that $\delta n_{i\vec{p}}$ is nonzero only for $p \approx p_F$ as $T \rightarrow 0$ [Eqs. (20) and (2)], we can evaluate this sum, substitute the result into Eq. (20), and then sum over \vec{p} and σ , leading to

$$\delta \mu_i = \frac{1}{\nu_i} [(1 + \nu_i s_{f_0}^{ii}) \delta n_i + s_{f_0}^{ij} \nu_j \delta n_j], \quad j \neq i \quad (22)$$

from which we are able to identify the derivatives in Eq. (19). Thus,

$$K_T = \left[n^2 V \left(\frac{1}{\nu_1} (1 + \nu_1 s_{f_0}^{11}) + 2 s_{f_0}^{12} + \frac{1}{\nu_2} (1 + \nu_2 s_{f_0}^{22}) \right) \right]^{-1}, \quad (23)$$

$$T \ll \min(T_{F_1}, T_{F_2})$$

K_T is thus temperature independent for low T . The bounds for the $s_{f_0}^{ij}$, Eqs. (15), are readily shown to correspond to the statement of positivity of the formal result for K_T .

It is interesting to relate the partial structure factors $S_{ij}(k)$ to Landau parameters. The $S_{ij}(k)$ are defined by

$$S_{ij}(k) = \delta_{ij} + (n_i n_j)^{1/2} \int d^3 r e^{i\vec{k}\cdot\vec{r}} [g_{ij}(\vec{r}) - 1], \quad (24)$$

where the $g_{ij}(r)$ are the pair distribution functions. For $k \rightarrow 0^+$, the structure factors are related to

the mean-square fluctuations in number density²³:

$$S_{ij}(k \rightarrow 0^+) = (\overline{N_i N_j} - N_i N_j) / (N_i N_j)^{1/2}, \quad i, j = 1, 2 \quad (25)$$

where the bar denotes thermal average. One can then show that

$$\frac{\partial \mu_i}{\partial N_j} \Big|_{N_j, \nu, T} = \frac{k_B T (N_i N_j)^{1/2} S_{i', j'}(0^+)}{N_1 N_2 [S_{11}(0^+) S_{22}(0^+) - S_{12}^2(0^+)]}, \quad (26)$$

where i' denotes the index complementary to i . For the case of a neutral two- (charged) component system (with $n_1 = n_2$, $e_1 = -e_2$) the requirement of electroneutrality then leads to the following relations, valid²⁴ for small k :

$$S_{ij}(k) = S(0^+) + a_{ij} k^2, \quad i, j = 1, 2 \quad (27)$$

where $S(0^+) = S_{11}(0^+) = S_{12}(0^+) = S_{22}(0^+)$ and where the a_{ij} cannot be determined solely on the basis of the neutrality requirement.

Substituting Eq. (27) into Eq. (26) we arrive at

$$\frac{\partial \mu_i}{\partial N_j} \Big|_{N_j, \nu, T} = \lim_{k \rightarrow 0^+} \left(\frac{1}{k^2 (a_{11} + a_{22} - 2a_{12})} + \frac{a_{i', j'}}{S(0^+) (a_{11} + a_{22} - 2a_{12})} \right). \quad (28)$$

We then see explicitly the divergences in the individual $\partial \mu_i / \partial N_j$'s but we also note the direct cancellation of these divergent parts in the expression for the compressibility. From Eqs. (22) and (28) and the discussion after Eqs. (15) we can relate the well-behaved ("screened") $s_{f_0}^{ij}$'s to the a_{ij} 's and $S(0^+)$:

$$s_{f_0}^{ij} = \frac{1}{\nu_i} \left(\frac{\nu_i a_{i', j'}}{S(0^+) (a_{11} + a_{22} - 2a_{12})} - \delta_{ij} \right), \quad i, j = 1, 2. \quad (29)$$

The isobaric thermal expansion coefficient

$$\alpha_p \equiv \frac{1}{V} \frac{\partial V}{\partial T} \Big|_{P, N_1, N_2}$$

for a general two-component system can be expressed most conveniently as

$$\alpha_p = \frac{K_T}{V} \left(S + N_1 \frac{\partial \mu_1}{\partial T} \Big|_{\nu, N_1, N_2} + N_2 \frac{\partial \mu_2}{\partial T} \Big|_{\nu, N_1, N_2} \right). \quad (30)$$

Now $\mu_i = \partial F / \partial n_i \Big|_{T, n_i}$, where $F = E - TS$. Then $\delta F = -S \delta T$, so using $S = C_V$ for low T and Eq. (18) we find for a general two-component Fermi liquid at low temperature:

$$F = F_0 - \frac{\pi^2}{4} k_B T^2 \left(\frac{N_1}{T_{F_1}^*} + \frac{N_2}{T_{F_2}^*} \right),$$

where F_0 is the ground state free energy and where

$$T_{F_i}^* = (9\pi^4)^{1/3} n_i^{2/3} / (2k_B m_i^* [n_1, n_2]).$$

The chemical potentials are then given by

$$\mu_i(n_1, n_2, T) = \mu_i(n_1, n_2, T=0)$$

$$- \frac{\pi^2}{4} k_B T^2 \left(\frac{1}{3T_{F_i}^*} + \frac{n_i}{m_i^* T_{F_i}^*} \frac{\partial m_i^*}{\partial n_i} - \frac{n_j}{m_j^* T_{F_j}^*} \frac{\partial m_j^*}{\partial n_i} \right),$$

$j \neq i, \quad T \ll \min(T_{F_1}, T_{F_2}). \quad (31)$

For a system with charged components ($e_1 = -e_2$), Eq. (31) is meaningful only when $n_1 = n_2$. Using Eq. (31) in Eq. (30) we then have

$$\alpha_p = K_T C_V \left[\frac{2}{3} - \frac{n}{m_1^* + m_2^*} \left(\frac{\partial m_1^*}{\partial n} + \frac{\partial m_2^*}{\partial n} \right) \right], \quad (32)$$

where expressions (23) and (18) for K_T and C_V must be used. We note that α_p varies linearly with T at low T , as expected.

Finally we obtain the phenomenological expression for the low-temperature weak-field static spin susceptibility $\chi = \partial M / \partial H$, where M is the magnetization density in the presence of the field H , [$M = \frac{1}{2} \hbar \gamma_1 (n_{1\uparrow} - n_{1\downarrow}) + \frac{1}{2} \hbar \gamma_2 (n_{2\uparrow} - n_{2\downarrow})$, where $\gamma_i \equiv g_i \mu_{B_i}$ is the gyromagnetic ratio with g_i the Landé g factor and $\mu_{B_i} = e \hbar / 2m_i c$ is the appropriate Bohr magneton.] We again start with Eq. (20) where now $\delta n_{i\vec{\sigma}}$ corresponds to the change upon addition of the small magnetic field. It can be shown that $\delta \mu \sim H^2$ in this case and we may neglect it to lowest order. The change in qp energy arises from both the direct "Zeeman" interaction and from the self-consistent change in the qp distribution functions due to the presence of the fields. Noting that in this case $\delta n_{i\vec{\sigma}} = \sigma \delta n_{i\vec{\sigma}}$ and using the fact that the spin-antisymmetric interspecies f functions vanish [Eq. (7)] we have

$$\delta \epsilon_{i\vec{\sigma}} = h_i \sigma + 2 \sum_{\vec{y}} a_{\vec{y}\vec{y}}^{f_{i\vec{y}}} \delta n_{i\vec{y}}, \quad (33)$$

where $h_i = -\frac{1}{2} \hbar \gamma_i H$. Using again $\delta n_{i\vec{y}} \sim \delta(\epsilon_{i\vec{y}} - \mu_i)$, we perform the sum in Eq. (33), substitute into Eq. (20), and then sum over \vec{p}, σ . This leads to

$$\chi = \frac{1}{4} \left(\frac{\nu_1 \gamma_1^2}{1 + \nu_1 a_{f_0^{11}}} + \frac{\nu_2 \gamma_2^2}{1 + \nu_2 a_{f_0^{22}}} \right), \quad T \ll \min(T_{F_1}, T_{F_2}) \quad (34)$$

which is T independent. The bounds for the $a_{f_0^{ii}}$'s, Eq. (15a), are equivalent to the statement that the formal result for χ for the normal Fermi-liquid Eq. (34) be positive. As $1 + \nu_i a_{f_0^{ii}}$ approaches zero from above, increasingly large long-wavelength fluctuations of the magnetic moment occur and χ diverges, signifying a transition to a magnetically ordered state.

Among the above expressions for C_V , K_T , α_p , and χ appear a total of nine Landau parameters (including the $\partial m_i / \partial n$). Unlike the case for one component, however, a complete empirical determination of these parameters is not possible. This is because that "partial" compressibility, spin susceptibility, and thermal expansion or specific heat measurements cannot be performed owing to, respectively, the requirement of charge neutrality, the nonexistence of "selective" magnetic fields coupling to only one species, and the inability to selectively heat one of the species. In any case the phenomenological description for the two component case is still fruitful: The "exact" temperature dependence of equilibrium properties as $T \rightarrow 0$ is given and these properties are related in a common framework. Moreover, for the case of LMH it will be possible to effectively reduce the number of relevant parameters by exploiting the large mass ratio of the two species.

IV. EQUILIBRIUM PROPERTIES OF NORMAL LIQUID METALLIC HYDROGEN

We now examine the above results when applied to the case of normal liquid metallic hydrogen, i.e., when the two species are protons (p) and electrons (e). As noted earlier, we have in mind a density corresponding to $r_s \simeq 1.6$ and temperatures $T \ll T_{F_p} \sim 10^2$ K.

We first estimate the order of magnitudes of the relevant Fermi-liquid parameters. Starting with the electrons, it is at once plausible that the replacement of the uniform positive background of the jellium model by a liquid of protons will not affect the electron qp-qp interaction energy by more than a factor of order unity. Thus we estimate $f^{ee} \sim f^J$ or perhaps more accurately, $f^{ee} \sim \tilde{f}^J$, where f^J is the electron f function in the jellium approximation and where ${}^{25} \tilde{f}^J(r_s) = f^J((\tilde{m}/m)r_s)$ is the f function for a rescaled jellium model. Here the bare mass m is replaced by a rescaled "bare" mass \tilde{m} chosen to include the effect of the electron-proton interaction in the original problem. We expect $\tilde{m} \sim m$.

Calculations of the f_i^J in the range of metallic densities have been carried out by a number of workers. Exemplary results are those of Rice,²⁶ who carried out the calculation in the Hubbard ap-

proximation. These results are in fact more justifiably applied to a high electron density metal like metallic hydrogen ($r_s \approx 1.6$) than to most other metals (to which they are regularly and with some accuracy applied). Defining the usual dimensionless parameters $F_i^{ee} \equiv \nu_e f_i^{ee}$ we expect from Rice's results that ${}^s F_0^{ee}$, ${}^a F_0^{ee} \sim 10^{-1}$, ${}^s F_1^{ee} \sim 10^{-2} - 10^{-1}$ for values of the parameter $\tilde{m} r_s$ in the range $\sim 0.5 - 5$.

Considering the protons next, we observe that we may take the point of view that LMH is an effective "one-component" liquid of appropriately dressed protons interacting with screened short-range forces. Effective proton pseudopotentials have been calculated and found to be for our purposes adequately modeled by a Yukawa form.⁶ They may also be crudely fitted to a Lennard-Jones-type interaction with a choice of a hard core diameter approximately half the interprotonic separation, as in the case of liquid ${}^3\text{He}$. In ${}^3\text{He}$ the interparticle separation is over twice that in LMH, but of course the atomic mass is 3 times larger; thus relative to ${}^3\text{He}$ the higher zero-point motion of the proton in LMH will, in fact, tend to "compensate" for the higher confinement. In fact, it has been estimated that the proton liquid in LMH has a deBoer quantum parameter about $\frac{2}{3}$ that of ${}^3\text{He}$.⁵

It thus appears that the ratio of mean potential energy to mean kinetic energy (of which, crudely speaking, the f 's are a measure) for the protons in LMH is comparable to (though probably higher than) that for the atoms of ${}^3\text{He}$. We therefore expect, in view of the values of the Landau parameters for ${}^3\text{He}$ taken from experiment,¹⁷ that the dimensionless parameters $F_l^{pp} \equiv \nu_p f_l^{pp}$ would be of the order of 1–10 (for $l=0$ and 1).

It remains to consider the interspecies f^{ep} , which corresponds to the interaction energy of an electron qp and a proton qp at the Fermi surface. Given the equal magnitudes of the bare interactions among all the particles, the similarity of the screening of these interactions and the similarity of the relative kinetic energies of a proton-electron pair and an electron-electron pair, we would expect the full f^{ep} to be comparable to the full f^{ee} . However, it is highly unlikely that this comparability can hold order by order in the respective Legendre expansions, for if we were to assume, e.g., ${}^s f_1^{ep} \sim {}^s f_1^{ee} \sim (0.01 - 0.1)/\nu_e$ in Eq. (13) (for $i=e$), we would be forced to conclude that the electron effective mass would be absurdly small, on the order of $\sim (0.01 - 0.1) m_e/m_p$. This problem is resolved, however, when it is realized that since m_p^*/m_e^* is so large, the energy of interaction of an electron qp of momentum \vec{p}_e ($|\vec{p}_e| = p_e$) and a proton qp of momentum \vec{p}_p ($|\vec{p}_p| = p_p$) cannot significantly depend upon the angle θ be-

tween \vec{p}_e and \vec{p}_p , i.e., regardless of θ , the proton is still essentially "fixed." Thus if one expands f^{ep} in a Legendre series in $\cos\theta$, clearly only the $l=0$ (i.e., isotropic) term is significant. In fact, using the relationship between Landau parameters and qp forward scattering amplitudes, one can show²⁷ that $f_i^{ep} \sim (m_e/m_p)^l$. Thus we have $f^{ep} \sim f_0^{ep} \sim f_0^{ee} \sim (0.1)/\nu_e$ while $f_1^{ep} \sim (m_e/m_p) f_1^{ee} \sim (m_e/m_p) (0.01 - 0.1)/\nu_e$. Note that this correct reduced magnitude of ${}^s f_1^{ep}$ guarantees that both m_p^*/m_p and m_p^*/m_e will be of order unity.

We now use these order-of-magnitude estimates of the Landau parameters in expressions (18), (23), (32), and (34) to discuss the low-temperature equilibrium properties of normal LMH. Using $m_e^* \gg m_e$ we find for the specific heat

$$C_V^{\text{LMH}} \approx \frac{m_p^* k_B^2 p_f}{3\hbar^3} T, \quad T \ll T_{Fp} \quad (35)$$

We thus note that very interestingly the specific heat of the normal liquid metallic phase would be strikingly larger than that in the corresponding solid metallic phase of comparable density: In the solid at low temperatures C_V is dominated by an electronic contribution $C_{V\text{el}} \sim \tilde{m}_e T$ (with $\tilde{m}_e \sim m_e^*$), the protons giving a negligible phonon contribution²⁸ $\sim T^3$. Thus $C_V^{\text{LMH}}/C_V^{\text{SMH}} \sim m_p^*/m_e^* \sim 10^3$ (SMH denotes solid metallic hydrogen.)

This difference is understood by noting that at a given temperature well below both T_{Fe} and T_{Fp} the proton qp distribution function as function of momentum \vec{p} is more "smeared" out at $p \approx p_F$ than the electron qp distribution function, i.e., the protons are less degenerate. Thus the protons have (in the ratio m_p^*/m_e^*) more available phase space than the electrons and hence contribute more so to the entropy. We observe that in the liquid the phonons in the "proton fluid" give rise, as in the solid, to a $\sim T^3$ contribution to C_V . But the proton quasiparticle excitations (which do not occur in the solid) have much more available phase space (in the ratio T^2) than the collective phonon excitations.

For the compressibility we use $\nu_p \gg \nu_e$, $f_0^{ee} \sim f_0^{ep}$, and $f_0^{pp} \sim 10^2 (m_e^*/m_p^*) f_0^{ee} \sim 10^{-1} f_0^{ee}$.²⁶ Though we cannot be totally sure of this last estimate within a factor of ~ 10 , it might appear that we can neglect ${}^s f_0^{pp}$ in Eq. (27) for K_T , leaving

$$K_T^{\text{LMH}} \approx \left[n^2 V \left(\frac{\pi^2 \hbar^3}{m_e^* p_F V} + {}^s f_0^{ee} + 2 {}^s f_0^{ep} \right) \right]^{-1}, \quad T \ll T_{Fp}. \quad (36)$$

Thus we tentatively conclude that the electrons "dominate" the compressibility, not unlike the case with solid metals. The main formal difference between the expression for the liquid Eq. (36) and the Fermi-liquid theoretic expression

for jellium as a model of a solid is the appearance of the explicit electron-proton term f^{ep} in Eq. (36). In any case we expect no striking difference in the magnitudes of the compressibilities of the normal solid and normal liquid phases of hydrogen at comparable densities.

Turning to the expansion coefficient equation (32) we have

$$\alpha_p^{\text{LMH}} \approx K_T^{\text{LMH}} C_V^{\text{LMH}} \left(\frac{2}{3} - \frac{\partial(\ln m_p^*)}{\partial(\ln n)} \right), \quad T \ll T_{Fp}. \quad (37)$$

For the corresponding solid phase at low temperature, the electronic contribution ($\sim T$) again dominates the phonon contribution ($\sim T^3$): $\alpha_p^{\text{SMH}} \approx \frac{2}{3} K_T^{\text{SMH}} C_{V_{\text{el}}}^{\text{SMH}}$. But using the above conclusion $K_T^{\text{LMH}} \sim K_T^{\text{SMH}}$, we see that the expansion coefficient of the liquid is again enormously greater than in the solid: $\alpha_p^{\text{LMH}}/\alpha_p^{\text{SMH}} \sim m_p^*/m_e^*$.

Just as noteworthy as the size of α_p is the possibility that it may actually be negative. From Eq. (37) we see that this occurs when the logarithmic derivative of the proton effective mass with respect to density is sufficiently positive. In the expression for α_p for normal liquid ^3He , corresponding to the parenthetic factor in Eq. (37) is the factor $\frac{2}{3} - \partial(\ln m_{\text{He}}^*)/\partial(\ln n)$ which, in fact, is negative at typical densities.²⁹ Thus, to the extent that we have argued the screened protons in LMH are "comparable" to the ^3He atoms in liquid ^3He , the possibility that normal LMH may similarly exhibit a negative expansion coefficient at low T cannot be dismissed. A "physical" explanation for the negative α_p of strongly interacting Fermi fluids at low T was offered by Breuckner and Atkins.³⁰ Roughly, they argue that as T is lowered, the dominating tendency is for the particles to want to order in momentum space in accordance with the ideal Fermi-Dirac distribution. The particles "facilitate" this by moving further apart on average in order to reduce the interaction, thereby becoming more free-particle-like.

Finally, for the spin susceptibility we use $\nu_e/\nu_p \sim \gamma_p/\gamma_e = m_e/m_p \ll 1$ and $\nu_p^a f_0^{pp} \sim -1$, $\nu_e^a f_0^{ee} \sim -0.1$ (Ref. 26) in Eq. (34) to obtain

$$\chi^{\text{LMH}} \approx \frac{1}{4} \frac{\nu_e \gamma_e^2}{1 + \nu_e^a f_0^{ee}}, \quad T \ll T_{Fp} \quad (38)$$

i.e., the electrons completely dominate. This is comparable in magnitude to what we expect in the solid provided that we are above the ordering temperature (probably well below T_{Fp}) of the proton spins in the crystal. Let us note, however, that if $\nu_p^a f_0^{pp} \leq -1$, a value within the range of our

rough estimate, the denominator of the proton contribution to χ changes sign, signaling the onset of a ferromagnetic instability in the protons (i.e., the system would no longer be normal).

V. CONCLUSION

By making use of a generalization of Landau-Fermi liquid theory to the case of two charged, unequal mass, spin- $\frac{1}{2}$ components, we have seen that certain low temperature equilibrium properties of a possible normal liquid metallic phase of hydrogen will differ appreciably from the corresponding properties of a possible normal solid metallic phase. In particular, the specific heat and thermal expansion coefficient are vastly larger (by $\sim m_p/m_e$) in the liquid (both quantities varying as $\sim T$ in either phase) because of the presence in the liquid of proton quasiparticle excitations. Moreover, the possibility of a *negative* thermal expansion coefficient in the liquid makes an interesting and sharp contrast with the behavior of the solid. The spin susceptibility and compressibility are comparable in both phases though.

The two-component formalism discussed here is extended to the calculation of transport properties in a later paper. There, as here, our goals are to understand the qualitative low-temperature behavior of the normal liquid, and to contrast the corresponding properties of the assumed solid and liquid phases in order to provide experimenters with bases for discrimination between the two phases.³¹

Again, at this juncture the existence of a liquid phase under the conditions discussed is still only a possibility. Furthermore, assuming the existence of the liquid, its character, e.g., whether superconducting, normal, ferromagnetic, etc., is also not yet clear. The present work is however a starting point for an examination of this last issue: For example, first-principles microscopic calculations of the Fermi-liquid parameters as functions of γ_s may reveal the existence of magnetic or spatial order at the actual observed densities.³² Moreover, if the calculated parameters at the actual density are compatible with "normal" behavior one would have a fairly complete quantitative description of the equilibrium properties of the normal liquid phase.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the support of NASA through Grant No. NSG-7487 and thank Dr. C. Jayaprakash for pointing out the second work cited in Ref. 17.

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- ⁴For a Fermi liquid with Coulomb interactions, kinetic energy ($\sim n^{2/3}$), eventually dominates potential energy ($\sim n^{1/3}$) as the density increases. For liquid metallic hydrogen the density range of this "ultimate" liquid phase is enormously greater than that at the insulator-metal transition. Thus, we must allow the possibility of the solid being preferred in the intermediate-density regime, i.e., the liquid phase discussed here need not correspond to the ultimate high-density liquid phase.
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- ⁸Sudip Chakravarty and N. W. Ashcroft, *Phys. Rev. B* **18**, 4588 (1978).
- ⁹Other calculations are reviewed in Ref. 1.
- ¹⁰For a review of current experimental work see A. Ruoff, in *High Pressure and Low Temperature Physics*, edited by C. W. Chu and J. A. Woolam (Plenum, New York, 1978).
- ¹¹We note that in all likelihood the first statically maintained sample of metallic hydrogen will be microscopic and inaccessibly buried in the high pressure maintaining apparatus making a direct, say, shear-wave attenuation measurement, difficult to obtain.
- ¹²This is to be contrasted with another example of a "two-component" Fermi liquid, the electron gas consisting of spin-up and spin-down components. Naturally this "two-component" problem is qualitatively different from the considered problem (even if the masses of the two species in the latter were the same) in that it involves one (anticommuting) field while the considered problem involves two separately anticommuting fields which, however, cross commute with each other.
- ¹³Brovman *et al.* (Ref. 3) discuss the possibility of a stable ($T=0$) filamentary configuration, with liquidlike behavior in one direction.
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- ¹⁸All of the formal results of this section and the general forms of the equilibrium results given in the following section carry over directly to the case of short-range interactions. Differences stemming from long-range versus short-range interactions arise in transport phenomena and in number-density fluctuations as discussed in Sec. III.
- ¹⁹Dipole-dipole forces are neglected here; these may play a role in possible superfluidity.
- ²⁰The energy of interaction of two qp of different mass and of momentum p_1 and p_2 with, for example, $|p_1| > |p_2|$ must, in general, clearly change when the masses of the particles are switched; i.e., the magnitude of the relative velocity then changes.
- ²¹ $m_i^* > 0$, for otherwise the energy of the system is lowered by adding a qp outside the Fermi surface, which is contrary to the requirement that the chemical potentials $\mu_i = \epsilon_{iF\sigma}$ be the minimum energy change possible when a qp of type i is added to the system.
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- ²⁷This is most simply seen in an approximation in which the qp forward scattering amplitude is viewed as equal to an effective T matrix (appropriate to dressed particles interacting with dressed interaction), then expanding T in the usual series in terms of phase shifts, and finally expanding in powers of the (small) mass ratio. The "truncatability" of the Legendre series for f^{ep} would, in a field theoretic setting, correspond to the analog here of the Migdal theorem.
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- ³¹Also, the formalism discussed here may be of some use in studying other two-component Fermi systems, e.g., electron-hole drops.
- ³²This might be done by increasing r_s (starting from the high-density "gas") and determining the critical value of r_s at which the first of the stability bounds [Eqs. (15)] breaks down.