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Elementary Excitations of Bose-Condensed, Spin-Aligned Atomic Hydrogen

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Ground-state properties and elementary excitations of superfluid spin-aligned atomic
H are studied using methods adapted from the theory of liquid ^{4}He. The ground state is a
$k = 0$ condensate of the $|\downarrow$ (electron), $\uparrow$ (proton)$\rangle$ hyperfine state with a small admixture of
$|\uparrow\uparrow\rangle$ due to the hyperfine interaction. Excitations can also be labeled approximately by
their hyperfine states. $\varphi_{\downarrow\uparrow}(k)$ excitations are phononlike. $\varphi_{\downarrow\downarrow}(k)$ and $\varphi_{\uparrow\uparrow}(k)$ are free-
particle–like with energy gaps, and $\varphi_{\uparrow\downarrow}(k)$ excitations resemble spin waves.

Two ground-state hydrogen atoms, in a relative
spin triplet state, interact via a potential which
is qualitatively similar to, although slightly weak-
er than, that of helium. When a collection of hy-
drogen atoms is forced into a spin-aligned state
by the application of a very large magnetic field
at low temperature, the effective pair interaction
is again the triplet potential. This potential is
quite accurately known from the variational cal-
culations of Kolos and Wolniewicz.[1]

Since H atoms are very light bosons and since
the system is weakly interacting in the low-den-
sity limit, Bose condensation is expected to occur
at fairly high temperatures (e.g., 1°K at 1000 cm^3/
mole). Properties of perfectly spin-aligned,
Bose-condensed atomic H have been calculated
by Dugan and co-workers,[2] Stwalley and Nosanow,[3]
and Miller and Nosanow.[4] There is general agree-
ment among these authors concerning the equation
of state of the superfluid. In addition, they pre-
dict a fluid-solid transition at about 50 atm for
$T = 0$.

The stability of spin-aligned H in a finite field,
against spin flips and recombination transitions
to molecular states, is much less well under-
stood. Jones et al.[5] suggested that the nuclear
hyperfine interaction would seriously perturb the
relative spin alignment of the atoms and thus lead
to rapid recombination. Stwalley[6] claims that
spin-flip transition rates due to hyperfine inter-
actions are proportional to $\exp(-\mu H/kT)$ and can
thus be made negligibly small with available lab-
oratory fields and temperatures. Quite recently,
Berlinsky et al.[7] have shown that *solid* atomic H
is unstable, even at $T = 0$, for fields less than
about 1 megagauss because the spin-wave excita-
tions lie at negative energy and thus provide a di-
rect channel for decay of the magnetization.
These calculations suggest, however, that the
gas may be stable in available laboratory fields
at densities for which the Bose-condensation tem-
perature is easily accessible.

The purpose of this Letter is to present new
theoretical results concerning the ground-state
properties and elementary excitations of Bose-
condensed, spin-aligned atomic H in a large but

finite magnetic field. Such a magnetic Bose fluid is considerably more complicated than a spin-0 fluid such as helium because of the extra spin degrees of freedom of the electron and proton, and because of the spin dependence of the interactions. Nevertheless, standard methods from the theory of liquid ^{4}He can also be carried over to the hydrogen problem, and a great deal of information can be extracted using only elementary techniques.

The Hamiltonian for the system may be written as

$$\mathcal{H} = T + \mathcal{H}_Z + \mathcal{H}_{hf} + \mathcal{H}_D + \mathcal{H}_{exch}, \tag{1}$$

where T is the kinetic energy, and $\mathcal{H}_Z$ is the Zeeman interaction of electron and proton spins with the applied field,

$$\mathcal{H}_Z = H\sum_j (\mu_e S_j^z - \mu_p i_j^z), \tag{2}$$

with μ_e and μ_p the magnitudes of the electron and proton magnetic moments, $\vec{S}_j$ and $\vec{i}_j$ the electron and proton spins, and H the magnetic field which is directed along $\hat{z}$. $\mathcal{H}_{hf}$ is the hyperfine interaction which may be written as

$$\mathcal{H}_{hf} = a\sum_j \vec{i}_j \cdot \vec{S}_j = \mathcal{H}_{hf}^0 + \mathcal{H}_{hf}^1, \tag{3}$$

where the secular term $\mathcal{H}_{hf}^0$ is the part of $\mathcal{H}_{hf}$ which commutes with $\mathcal{H}_Z$ while the nonsecular term $\mathcal{H}_{hf}^1$ is the remainder, and $a = 0.047$ cm^{-1}.

The direct part of the atom-atom interaction is obtained by averaging the singlet and triplet pair potentials over spin states of the pair:

$$\mathcal{H}_D = \tfrac{1}{2}\sum_{i,j} V(r_{ij}), \tag{4}$$

with $V(r_{ij}) = \tfrac{1}{4}[V_S(r_{ij}) + 3V_T(r_{ij})]$. Similarly the exchange interaction is

$$\mathcal{H}_{exch} = \tfrac{1}{2}\sum_{i,j} J(r_{ij})\vec{S}_i \cdot \vec{S}_j, \tag{5}$$

where $J(r_{ij}) = V_T(r_{ij}) - V_S(r_{ij})$.

It is useful to begin by temporarily ignoring the nonsecular hyperfine interaction $\mathcal{H}_{hf}^1$. Then $\mathcal{H}_Z$ and $\mathcal{H}_{hf}^0$ favor the $|\downarrow(\text{electron}), \Downarrow(\text{proton})\rangle$ magnetic state. One approach to the problem, which is analogous to that employed by Feynman for ^{4}He,[8] is to assume a knowledge of the state φ_0 with energy E_0, which is an exact eigenstate of the Hamiltonian with all atoms in the $|\downarrow\Downarrow\rangle$ state. φ_0 is clearly not the ground state of the system since states containing molecules will lie below it. However φ_0 is a stationary state which may be metastable if small perturbations to φ_0 lie at positive energy with respect to E_0.

Appropriate trial wave functions for the low-ly-

ing excitations of the state φ_0 are of the form

$$\varphi_{\downarrow\uparrow}(r_1, \ldots, r_N) = \sum_j f_1(r_j)\varphi_0, \tag{6a}$$

$$\varphi_{\downarrow\Downarrow}(r_1, \ldots, r_N) = \sum_j f_2(r_j)i_j^-\varphi_0, \tag{6b}$$

$$\varphi_{\uparrow\Downarrow}(r_1, \ldots, r_N) = \sum_j f_3(r_j)i_j^- S_j^+\varphi_0, \tag{6c}$$

$$\varphi_{\uparrow\uparrow}(r_1, \ldots, r_N) = \sum_j f_4(r_j)S_j^+\varphi_0, \tag{6d}$$

and minimization of the average value of $\mathcal{H}$ (still taking $\mathcal{H}_{hf}^1 = 0$) with respect to these states leads to $f_I(r_j) = \exp(i\vec{k}\cdot\vec{r}_j)$, with $I = 1, \ldots, 4$.

The energies associated with these states are

$$\epsilon_1(k) = \hbar^2 k^2/2MS(k), \tag{7a}$$

$$\epsilon_2(k) = \hbar^2 k^2/2M + \mu_p H + \tfrac{1}{2}a, \tag{7b}$$

$$\epsilon_3(k) = \hbar^2 k^2/2M + (\mu_e + \mu_p)H - \tfrac{1}{2}\tilde{J}(0), \tag{7c}$$

$$\epsilon_4(k) = \hbar^2 k^2/2M + \mu_e H + \tfrac{1}{2}a + \tfrac{1}{2}[\tilde{J}(k) - \tilde{J}(0)], \tag{7d}$$

where $S(k)$ is the Fourier transform of the pair distribution function $g(r)$, which is obtained by integrating φ_0^2 over all coordinates except for the relative coordinate of a pair of atoms; and

$$\tilde{J}(k) = \rho \int J(r)g(r)e^{i\vec{k}\cdot\vec{r}} d^3r, \tag{8}$$

with ρ the density of the fluid. In the helium problem, $S(k)$ and hence $g(r)$ were known experimentally from neutron scattering data. However, for hydrogen one must rely on calculated distribution functions such those of Dugan and co-workers[2] or of Goldman.[9]

At this point several comments are in order. First, the wave functions defined by Eqs. (6) are of particular interest for this problem not only because they are good trial wave functions but also because they describe disturbances which could be induced, for example, by neutrons or by spatially varying magnetic fields. Second, the energies in Eqs. (7) may be interpreted as follows: $\epsilon_1(k)$ is phononlike since $S(k)$ must go like k in the long-wavelength limit. $\epsilon_2(k)$ is free-particle-like with a small gap at $k = 0$. $\epsilon_3(k)$ is also free-particle-like with a gap $(\mu_e + \mu_p)H - \tfrac{1}{2}\tilde{J}(0)$. Thus a minimal stability condition for the state φ_0 is that the cost in Zeeman energy of a spin flip be greater than the average gain in exchange energy. This condition was assumed to be true in Ref. 7 because an analogous condition applies in the solid. The exchange contribution to $\epsilon_4(k)$ is quite similar in form to the energy of a spin wave in a ferromagnet. Finally one notes that the assumed metastability of the state φ_0 depends crucially on the fact that the pair distribution function $g(r)$ goes to zero for small r much faster than $J(r)$ becomes large. Thus the integral in

Eq. (8) samples mainly the region just inside of the minimum of V_T and not the "molecular" region around $r = 0.75$ Å where $J(r) \simeq 10$ eV.

. The advantages of the variational approach are its simplicity and that it leads to the very important result of Eq. (8). However, for higher-order corrections to the wave functions and, particularly, for more complicated interactions such as $\mathcal{K}_{hf}^{1}$, the method quickly becomes cumbersome. In addition, the trial wave functions of Eq. (6) appear rather ad hoc, and hence from a pedagogical point of view it is useful to show that Eqs. (7) can also be derived in a somewhat different way.

To do this, the Hamiltonian is written in a second-quantized form in terms of atom creation operators $C_{m,\mu}^{\dagger}(k)$, where $m, \mu = \pm 1$ are the magnetic quantum numbers of the electron and proton spins, respectively. Then, following Bogoliubov,[10] one assumes that the lowest-lying $k = 0$ state is

macroscopically occupied; i.e., for an N atom system,

$$\langle C_{-1,1}{}^{\dagger}(0)\rangle = \langle C_{-1,1}(0)\rangle = \sqrt{N_0} \simeq \sqrt{N}. \tag{9}$$

The presence of $\mathcal{K}_{hf}^{1}$ leads to a term linear in $C_{1,-1}{}^{\dagger}(0)$. Thus the $k = C$, $|\uparrow\downarrow\rangle$ state is also macroscopically occupied. That is, one must write

$$\langle C_{1,-1}{}^{\dagger}(0)\rangle = \langle C_{1,-1}(0)\rangle = \gamma\sqrt{N_0}, \tag{10}$$

and it will be shown that, for large fields, γ is proportional to a/H. The unknown constant N_0 can be eliminated by use of the relation

$$(1 + \gamma^2)N_0 = N - \sum_{k,m,\mu}{}' C_{m\mu}{}^{\dagger}(k)C_{m\mu}(k), \tag{11}$$

where the prime over the summation means that the terms with $k = 0$ and $\mu = -m$ are excluded. Equations (9)–(11) lead to an effective Hamiltonian of the form

$$\mathcal{K} = E_0(\gamma) + \sum_{k,m,\mu}{}' \epsilon_{m\mu}(k)C_{m\mu}{}^{\dagger}(k)C_{m\mu}(k) + \sum_{k,m,n}{}' \{[M(m,n;k) + \tfrac{1}{2}a\delta_{m,-n}]C_{m,-m}{}^{\dagger}(k)C_{n,-n}(k)$$

$$+ \tfrac{1}{2}M(m,n;k)[C_{m,-m}{}^{\dagger}(k)C_{n,-n}{}^{\dagger}(-k) + C_{m,-m}(k)C_{n,-n}(k)]\}$$

$$+ \frac{\gamma}{4(1+\gamma^2)}\sum_{k,m}{}' J(k)[C_{mm}{}^{\dagger}(k)C_{-m,-m}{}^{\dagger}(-k) + C_{mm}(k)C_{-m,-m}(-k)], \tag{12}$$

plus cubic and higher-order terms in the $C_{\mu m}{}^{\dagger}(k)$, where

$$N^{-1}E_0(\gamma) = -\tfrac{1}{2}H(\mu_e + \mu_p)\left(\frac{1-\gamma^2}{1+\gamma^2}\right) - \frac{a}{4} + \frac{a\gamma}{1+\gamma^2} + \tfrac{1}{2}V(0) + \tfrac{1}{8}J(0)\left(\frac{1-\gamma^2}{1+\gamma^2}\right)^2, \tag{13a}$$

$$\epsilon_{m\mu}(k) = \frac{\hbar^2 k^2}{2M} + \mu_e H\delta_{m,1} + \mu_p H\delta_{m,-1} - \frac{\gamma^2}{1+\gamma^2}(\mu_e + \mu_p)H + \tfrac{1}{2}a\delta_{m,\mu} - \frac{a\gamma}{1+\gamma^2}$$

$$- \tfrac{1}{4}J(0)\left[\left(\frac{1-\gamma^2}{1+\gamma^2}\right)^2 + m\frac{1-\gamma^2}{1+\gamma^2}\right] + \frac{J(k)}{2(1+\gamma^2)}(\delta_{m,1} + \gamma^2\delta_{m,-1})\delta_{m\mu}, \tag{13b}$$

$$M(m,n;k) = \frac{V(k)}{1+\gamma^2}(\delta_{m,-1} + \gamma\delta_{m,1})(\delta_{n,-1} + \gamma\delta_{n,1}) + \frac{J(k)}{4(1+\gamma^2)}(\delta_{m,-1} - \gamma\delta_{m,1})(\delta_{n,-1} - \gamma\delta_{n,1}). \tag{13c}$$

The excitation energies for a general quadratic Hamiltonian such as this are most easily found by use of a matrix Green's-function technique.[11] In this case the matrices are 8×8 since there are four kinds of particles for which particle number is not conserved. However, the matrices reduce to two 4×4 blocks since $L = |m + \mu|$ is a good quantum number. In fact, for $L = 1$, the Green's function reduces even further to 2×2 blocks, and the excitation energies which result are essentially identical to $\epsilon_2(k)$ and $\epsilon_4(k)$, with $\tilde{J}(k)$ replaced by $J(k)$, provided that γ is sufficiently small, as will be discussed below.

The more interesting case is $L = 0$ for which the

inverse Green's function is a 4×4 matrix. Matrix inversion leads to a fourth-order secular equation which can be solved exactly for the squares of the excitation energies, z^2, as functions of γ and the various interaction parameters. When the smaller of the two roots is expanded as a power series in the small quantities γ and $a/|\epsilon_{1,-1}(k) - \epsilon_{-1,1}(k)|$, one obtains, for $k = 0$,

$$z^2 = 2[V(0) + \tfrac{1}{4}J(0)][\epsilon_{-1,1} - a^2/4\epsilon_{1,-1}], \tag{14}$$

plus terms of order γ^3. The first square bracket is simply the $k = 0$ Fourier transform of the triplet potential, which is certainly positive. Using

Eq. (13b) one finds that the second bracket has the absolute value

$$-\gamma a - \gamma^2 \Delta - a^2/4\Delta, \tag{15}$$

where

$$\Delta = \epsilon_{1,-1} = (\mu_e + \mu_p)H - \tfrac{1}{2}J(0), \tag{16}$$

again ignoring terms of order γ^3. To this level of accuracy, the value of γ is simply that which minimizes $E_0(\gamma)$ in Eq. (13a). This gives $\gamma = -a/2\Delta$ which is also the value of γ for which Eq. (15), and hence the lowest excitation energy, is zero. With further algebraic manipulations, one finds that for small k the energy is linear in k, as is appropriate for a phononlike mode.

These conclusions could certainly be modified slightly by terms which are of higher order in γ; but given the complexity of the algebra and the other uncertainties in the theory, a higher-order calculation is not really warranted. The important result is that for large magnetic fields the nonsecular part of the hyperfine interaction, $\mathcal{K}_{hf}{}^1$, does not lead to any gross instabilities but instead simply modifies the ground-state wave function in a perturbative way.

Finally it is worth considering for a moment the behavior of superfluid H in an inhomogeneous magnetic field. If all electron spins are antiparallel to the field, then it is clearly advantageous for the atoms to move to the high-field region until some equilibrium density is reached at which the nonmagnetic pressure equals the gain in energy density from the magnetic field. This magnetostrictive effect can be used as a method for collecting spin-aligned hydrogen. The density which can be achieved for a given field can easily be estimated using the equations of state which have been calculated for the triplet potential. It is important to keep in mind, however, that in order for the superfluid to be stable against spin flips, the quantity Δ in Eq. (16) [or more accurately, $\tilde{\Delta}$, which is obtained by replacing $J(0)$ in Eq. (16) by the $\tilde{J}(0)$ from Eq. (8)] must be positive and large compared to the hyperfine constant a.

This gives rise to an independent relationship between the field and the density which is determined mainly by the exchange interaction. A comparison of these two relationships shows that a localized field in equilibrium with a particle source will always collect a sufficiently high density of particles to drive Δ negative. Thus, as a practical matter it would seem necessary to limit the supply of H atoms if the superfluid is to remain stable.

The next logical step in the theory of superfluid hydrogen is to treat the two-excitation–scattering problem, since the two-particle states are connected to the single-particle states by cubic terms in the Hamiltonian, which I have neglected, and since two-particle interactions could easily lead to a fundamental instability toward molecule formation. Such calculations are in progress, and in the meantime it is hoped that the theory presented here will be of some assistance to experimentalists who are working to produce this very exotic new material.

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