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BOSE CONDENSATION IN SPIN POLARIZED ATOMIC HYDROGEN

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Résumé.- Nous proposons une description phénoménologique de l'hydrogène polarisé suggérée par l'hamiltonien microscopique et qui inclut deux particules de Bose qui correspondent, à la limite des faibles densités, aux deux états hyperfins les plus bas. On s'attend à ce que l'expérience peuplé initialement les deux états et à ce que les temps d'équilibre soient longs. Quand la condensation de Bose a lieu dans chacun des deux états, il apparaît une aimantation cohérente spontanée perpendiculairement au champ de stabilisation ; ceci devrait être facilement observable par une simple expérience de résonance magnétique. Les inhomogénéités de champ ne devraient pas élargir le signal de résonance dans une telle expérience, contrairement à ce qui se passe avec des systèmes non condensés.

Abstract.- A phenomenological description of spin-polarized hydrogen, suggested by the microscopic Hamiltonian, is proposed that includes two Bose particles which correspond in the low density limit to the two lowest hyperfine states. Experiments are expected to initially populate both states, and equilibration times will be long. When Bose condensation occurs in both states a spontaneous coherent magnetization perpendicular to the stabilizing field will appear ; that should be readily observable in a simple magnetic resonance experiment. Field inhomogeneities should not broaden the resonance signal in such an experiment, contrary to one's experience with uncondensed systems.

Should it prove possible to stabilize spin polarized atomic hydrogen, $H\uparrow$, at densities, ($\sim 10^{19} \text{ cm}^{-3}$), high enough to observe Bose Condensation, the question of how to unambiguously detect the condensed phases will arise. Atomic hydrogen will be at best metastable in any attainable laboratory field and recombination rates are very difficult to anticipate. Thus, the canonical He^4 superflow experiments may not be possible. Our attention has focused on the magnetic properties of the superfluid phases of $H\uparrow$ which in many respects are expected to resemble those of a spin 1/2 Boson (i.e. a Boson with two internal states). The stabilizing field is ready made for a magnetic resonance experiment that is non-invasive and can be done on a much shorter time scale than the traditional superfluid flow experiments. We will describe one effect which is unique to superfluid $H\uparrow$ and whose magnitude should render it easily measurable.

It is expected that a magnetic field can be used to select just the two lowest hyperfine states of atomic hydrogen $/1/$. The upper two, whose electron spin is predominately parallel to the field, will be rejected from the experimental region. In

a field, we will assume hereforth of order 100 kG, the spin dependence of the low energy states is

$$\begin{aligned} |a\rangle &= (| - +\rangle - \epsilon | + -\rangle) / \sqrt{1+\epsilon^2} \\ |b\rangle &= | - -\rangle \end{aligned} \tag{1}$$

where within each ket the signs refer to the direction of electron and proton spins respectively.

The parameter ϵ is the ratio of the hyperfine constant, a , to the Zeeman energy $2\mu_e H_z$ and is of order 2×10^{-3} . Also, $E_b - E_a \sim 50 \text{ mK}$. When the density in atomic units is low, and translational energies less than atomic excitation energies we can treat states a and b as Bosons. The corresponding second quantized field operators will be denoted by ψ_a, ψ_b .

Most of the physics we will discuss follows from the Hamiltonian:

$$H = H_0 + \frac{1}{2} \sum_{ij} V_D(r_{ij}) + \frac{1}{2} \sum_{ij} V_E(r_{ij}) S_i \cdot S_j \tag{2}$$

where H_0 includes the kinetic and intraatomic energies as well as the interaction with the external field $/2/$. The electron spin is denoted by \tilde{S} and $V_D = \frac{1}{4} (V_S + 3V_T)$, $V_E = V_T - V_S$, where V_T and V_S are the well known triplet and singlet potentials between hydrogen atoms.

We can rewrite (2) within the space of states ψ_a, ψ_b in the form presented in (3) below. (A more careful discussion of the effects of coupling to the upper hyperfine states is contained in Reference /3/. See also /2/.)

$$H = H_0 + \int \frac{1}{2} (\rho_a + \rho_b) (1) (V_T - \frac{\epsilon^2}{2} V_E) (1-2) (\rho_a + \rho_b) (2) - \frac{\epsilon^2}{4} \left\{ \rho_a(1) V_E(1-2) \rho_a(2) - \rho_b(1) V_E(1-2) \rho_b(2) \right\} + \frac{\epsilon^2}{4} \left\{ S_+(1) V_E(1-2) S_-(2) + S_-(1) V_E(1-2) S_+(2) \right\} + \mathcal{O}(\epsilon^3), \quad (3)$$

where the operators $\rho_{a,b} = \psi_a^+ \psi_a, \psi_b^+ \psi_b$ and $S_{\pm} = S_x \pm iS_y = \psi_a^+ \psi_b, \psi_b^+ \psi_a$. Numerical arguments are integrated over all space. Along with (3), we can write the magnetization density, m_i , as

$$m_{x,y} = \mu_{\pm} S_{x,y} \\ m_z = \mu_z S_z \quad (4a)$$

with $S_z = \frac{1}{2} (\rho_a - \rho_b)$ and

$$\mu_{x,y} \equiv \mu_{\pm} = (\mu_p + \epsilon \mu_e) \\ \mu_z = (\mu_p - \epsilon^2 \mu_e) \quad (4b)$$

We have neglected in m_z a background piece proportional to the total density.

There is an important separation of time scales implicit in (3). The thermalization time T_{th} of the center of mass motions is set by the largest term in (3), $(V_T - \frac{\epsilon^2}{2} V_E)$. The last term in (3) acts like an antiferromagnetic exchange in the x-y direction and commutes with the integral of S_z but not S_x or S_y . It thus contributes to the transverse magnetization relaxation time, T_2 . Finally the dipolar interactions we have omitted from (3) fail to commute with all the components of the spin and enter both the relaxation time, T_1 of the relative populations $\rho_a - \rho_b$ and the transverse relaxation time T_2 . At low temperatures we expect

$$T_{th} \ll T_2 \ll T_1$$

For the time being, let us take the T_1 due to atomic processes infinite in comparison to T_{th} .

Then as a mixture of H⁺ in the states a and b with

densities n_a, n_b is cooled, we expect Bose condensation to occur separately in the two states at temperatures set by the ideal Bose gas formulae. In thermodynamic equilibrium Bose condensation is characterized by an order parameter $\langle \psi \rangle = \psi_0 e^{i\phi}$, ($\psi_0 = |\langle \psi \rangle|$), that is complex and uniform in space. If our ensemble corresponds to long range order in both states a and b the expectation value of the operators $m_{x,y}$ in (4) representing the perpendicular magnetization density will have a non-zero magnitude very nearly equal:

$$\sigma_{\perp} = \mu_{\pm} \psi_0^a \psi_0^b \quad (5)$$

and a direction in the x,y plane set by $\phi_a - \phi_b$.

(Whether one regards the state implicit in (5) as having two condensates or a single condensate formed by superimposing the two "spin" states a,b is a moot point.)

If the relative populations of a to b are not themselves in equilibrium, the perpendicular magnetization σ_{\perp} will rotate at a rate

$$\nu_{a,b} = (\nu_a - \nu_b)/h \sim 10^9 \text{ hz} \quad (6)$$

where $\nu_{a,b}$ are the chemical potentials of hydrogen atoms in the two hyperfine levels a,b. (Microscopic considerations suggest quite strongly that one can not have Bose condensation in both states a and b and $\mu_a = \mu_b$.) This is the effect to which we alluded in the Introduction. A sample of 10^{19} coherently rotating spins with a magnetic moment several times the proton's should be observable in a magnetic resonance experiment.

A number of complications have to be addressed before we can assert that the above effect will be measurable /3/.

1. Inhomogeneous Broadening

Superfluid H⁺ is a rather interesting system in this respect since the signal, (6), appears as the difference of two chemical potentials. The chemical potential must be uniform in any state in

mechanical equilibrium. A field inhomogeneity will induce a pressure gradient and ultimately lead to uniform values of μ_a, μ_b . Thus, we expect no inhomogeneous broadening of the line (6).

2. Relaxation Times

Initially a mixture of hydrogen atoms in the a, b states will have a zero value of σ_{\perp} . When such a mixture is quenched to below the Bose condensation temperatures of $n_{a,b}$ what are the time scales of the kinetic processes that lead to the appearance of σ_{\perp} in (5)? Are they of order the atomic T_2 times or because σ_{\perp} is tied to the order parameter will it develop on a time scale set by V_T (e.g. T_{th}) in (3)? We have addressed this question by doing a microscopic calculation to fix the parameters in a mode coupling Ginzburg-Landau theory. The magnitude of the order parameter should relax to a constant uniform value in a time of order 10^{-8} sec. The phase of the order parameter, and thus σ_{\perp} , evolves diffusively with a diffusion constant of order 10^{-3} cm²/sec. In a time measured in seconds, millimeter size domains of σ_{\perp} will develop. In each domain, σ_{\perp} rotates uniformly at a rate given by (6). Its average over the entire system is zero until effects that break the conservation of $S_{x,y}$ come into play. The subsequent evolution of σ_{\perp} is determined by the magnitude of T_2 and the long range dipolar forces.

3. Domain Energetics

When macroscopic regions of nearly uniform σ_{\perp} develop, we must enquire as to what limits the domain size. Domain formation is favored by the small dipole fields of order $4\pi\mu_{\perp}n \sim 10^{-3}$ G (for $n \sim 10^{19}$ cm⁻³). Normally, domain sizes are set by balancing exchange against dipolar energies with the thickness of domain walls determined by the anisotropy energy. There is no intrinsic aniso-

tropy energy in H_† so σ_{\perp} will vary smoothly in space. The only analogue of the exchange energy is the superfluid density that acts as a stiffness constant against changes in ϕ_a or ϕ_b . An order of magnitude estimate yields domains of size ~ 0.1 cm.

A more precise calculation has been done for a long cylinder of radius R, parallel to the stabilizing field with $\phi_a - \phi_b = kz$. (The phase varies linearly with z since although the superfluid velocity may be nonzero in thermodynamic equilibrium the divergence of the mass current must be zero; so if $\rho_s^{a,b}$ are uniform, $\nabla^2\phi_{a,b} = 0$). A precise calculation of the magnetostatic energy as a function of k gives

$$\frac{E}{V_0 l} \approx \frac{\hbar^2}{m} nk^2 + 4\pi(\mu_{\perp}n)^2 K_1(kR)I_1(kR) \quad (7)$$

and a minimum at $k = \frac{2\pi}{k} \approx .28$ cm for $n_a = n_b = n = 10^{19}$ and $R = 1$ cm. (Above K_1, I_1 are modified Bessel functions of the first order).

Actually this is still an underestimate of the domain size since we have not imposed the boundary conditions on v_s^a, v_s^b on the ends of the cylinder. The global flow field will be considerably more complicated than we have assumed and the introduction of vortices will probably be required in thermal equilibrium.

4. External Josephson Effect

We have chosen this term by analogy with Leggett's "internal Josephson Effect", which he used to explain the longitudinal resonance in superfluid He³-A /4/. In H_†, there appear to be no bulk energies, irrespective of size, that pick out a direction for $\phi_a - \phi_b$ even if it weren't rotating. The long range dipole energy will couple $\phi_a - \phi_b$ to the shape of the sample and if the resulting demagnetizing fields are inserted back into Bloch's equations, characteristic, though very small, harmonics of $v_{a,b}$ will appear. There will be a

corresponding small oscillation in the relative populations but no relaxation.

A more important source of coupling between the a-b populations is an external coil. If it is resonant, energy will oscillate between the field and sample. The coupling to the coil plays the same role here as the dipolar coupling did in Leggett's theory. The resistance of the coil will probably set the thermalization time of n_a/n_b as long as both phases are superfluid although real radiation damping is also possible.

We can now return to the question of whether the signal (6) will be measurable in Bose condensed H^+ . Let us assume we can stabilize a density of 10^{19} atoms for at least several seconds. Domains will form, σ_{\perp} locally will rotate at the same frequency (6), but the volume average of σ_{\perp} may be zero. If the frequency of a surrounding coil whose field exceeds μ_n is swept through $\nu_{a,b}$, the various domains will be pulled into alignment. If the Q of the coil is sufficiently high and if it remains locked on resonance, the external r.f. source can be turned off. The sample of hydrogen with its rotating magnetization will induce a sufficient field in the coil to keep the domains aligned.

One possible complication is that $\nu_{a,b}$ will change with $n_a - n_b$ in a calculable way. The resonant coil has therefore to track a variable frequency. In addition, populations should be changed sufficiently slowly so that the local equilibrium arguments we have used remain valid. The small ratio between the dipolar field and H_z insures all transverse r.f. sources can be kept small and rotation rate of the z component of the magnetization slow.

We have also investigated the hydrodynamics of superfluid H^+ and developed a suitable mode coupling theory that includes σ_{\perp} (which is not a

hydrodynamic variable) and reduces to Bloch's equations in the appropriate limit.

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A more complete list of references appears in /3/.

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