

Hydrodynamics of the condensed phases of spin-polarized atomic hydrogen

Eric D. Siggia and Andrei E. Ruckenstein

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853

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The thermodynamic, hydrodynamic, and kinetic properties of a variety of metastable states accessible to spin-polarized hydrogen are examined. The state with Bose condensation in the two lowest hyperfine levels will phase separate at a rate determined from superfluid hydrodynamics. The transverse and longitudinal magnetic relaxation times are evaluated in the classical, quantum, and superfluid regimes.

Atomic hydrogen in a large magnetic field (~ 100 kG), $H\downarrow$, is expected to behave as a weakly interacting Bose gas at low temperatures and densities $\leq 10^{19}$ cm^{-3} .¹⁻³ When the two lowest atomic hyperfine states are involved in the Bose condensation, $H\downarrow$ will resemble a superfluid spin- $\frac{1}{2}$ Bose gas.⁴ We intend here to clarify this analogy and explore those features of the $H\downarrow$ system, such as magnetic resonance, that are not shared by an ordinary spin-zero Bose gas.

Most experimentally interesting properties of condensed quantum systems can be understood by means of hydrodynamic arguments once the broken symmetry is identified thermodynamically. Even if the possibility of recombination to molecular hydrogen is put aside,^{5,6} the phases of $H\downarrow$ relevant to this article are only metastable. Metastable equilibria are not a problem if the various relaxation times are well separated so that the system passes from one near-equilibrium state to another of generally lower symmetry. This situation unfortunately does not prevail in $H\downarrow$ where a number of kinetic processes proceed at roughly similar rates. In addition, certain lifetimes

have proven difficult to estimate since the transformations involved resemble spinodal decomposition and cannot be calculated perturbatively around some uniform thermodynamic state.

These uncertainties and the desire to separate the well understood from the conjectured, have led us to consider the thermodynamics and hydrodynamics for the Hamiltonian describing $H\downarrow$ with certain small terms either neglected or adjusted to stabilize an otherwise unstable phase. We then proceed to estimate the relaxation times between the various phases accessible to $H\downarrow$ and conclude with a discussion of possible experiments to measure the new effects we have proposed.

In a large magnetic field it is convenient to work explicitly in terms of the two lowest atomic hyperfine states, $|a\rangle$ and $|b\rangle$, whose energy difference we define as H_z .⁴ Define a spin density in second quantized form, $\vec{S} = \frac{1}{2} \Psi^+ \vec{\tau} \Psi$, where τ_i are the Pauli matrices and $\Psi^+ = (\psi_a^+, \psi_b^+)$. We may write the total Hamiltonian as the sum of H_0 , H_1 , and H_d where for $H_z > 0$,

$$H_0 = \int \left(\frac{\hbar^2}{2m} |\nabla \Psi(x)|^2 - H_z S_z(x) \right) dx + \frac{1}{2} \int \int \rho(x) \left(V_T - \frac{\epsilon^2}{2} V_E \right) (x-x') \rho(x') dx dx' \quad (1a)$$

$$H_1 = \frac{\epsilon^2}{4} \int \int \{ [S_+(x) V_E(x-x') S_-(x') - \rho(x) V_E(x-x') S_z(x')] + \text{H.c.} \} dx dx' + O(\epsilon^3) \quad (1b)$$

and H_d is the dipolar energy. In (1), V_T and V_E are the triplet and exchange potentials, $\rho = \Psi^+ \Psi$, and ϵ is the amplitude of the small admixture of the electron spin-up state in $|a\rangle$. (We have neglected a small coupling of order ϵV_E between $|a\rangle$ and the other z spin-zero state, $|c\rangle$ in Ref. 4 which would be manifest at low temperatures by a shift in the effective g

factor for the spontaneous perpendicular magnetization.^{4,7})

The terms in H are grouped by symmetry. The dipole interaction relaxes S_z (rate T_1^{-1}) and $S_{x,y}$ (rate T_2^{-1}); H_1 also contributes to T_2^{-1} ; while the largest term, H_0 , thermalizes the kinetic energies (rate T_{th}^{-1}) and $T_{\text{th}} \ll T_2 \leq T_1$.

We begin by neglecting H_1 and H_d and retaining only H_0 . The thermodynamic ensemble is defined by the total energy, number density, and $\int \bar{S}(x)$. The imposed values of $S_z, S_{x,y}$ are enforced by the thermodynamic fields $h_z = \mu_a - \mu_b$ and \bar{h}_1 . Ensembles with a nonzero value of $S_{x,y}$ could be prepared by magnetic resonance.

The thermodynamic transitions in this model system are best understood in the frame with the z axis tilted to coincide with $\langle \bar{S} \rangle$ [itself parallel to $(h_1, h_z + H_z)$]. The first transition to occur as the temperature is lowered to $T_{\lambda a}$ is ordinary Bose condensation in the rotated spin state of higher density. At a lower temperature, $T_{\lambda b}$, the other spin state condenses and a component of the magnetization, $\bar{\sigma}_1$, perpendicular to the rotated z axis appears. Of course, since we have fixed $\langle \bar{S} \rangle$, domains will form in $\bar{\sigma}_1$ to keep its volume average zero.

An analog to the transition at $T_{\lambda b}$ occurs in a Heisenberg ferromagnet in a field with an imposed value of $\langle \bar{S} \rangle$. At sufficiently low temperatures, the thermodynamically preferred magnetization will exceed the imposed one in magnitude and a $\bar{\sigma}_1$ will develop spontaneously. In $H \downarrow$ as well as the magnet, it then follows that below $T_{\lambda b}$ the conjugate fields satisfy $\bar{h}_1 = 0$ and $h_z = -H_z$ and thus the free energy per volume for $H \downarrow$ becomes

$$F(N_a, N_b, T) = F_0(N_a + N_b, T) - H_z \langle S_z \rangle \quad (2)$$

(If $H_z + h_z \neq 0$, for instance, then the full magnetization would rotate into the z axis and exceed the imposed value.)

Below $T_{\lambda b}$, since $\bar{h}_1 = 0$ and $h_z = -H_z$, the hydrodynamics are those of a condensed spin- $\frac{1}{2}$ Bose gas with a superimposed rotation generated by H_z .⁸ In addition to first and second sound, there are two ferromagnetic spin-wave modes making four propagating modes in the linearized hydrodynamics.

Next let us retain H_1 in addition to H_0 so that $S_{x,y}$ are no longer conserved. Strictly speaking, properties of the phases so obtained are only relevant to $H \downarrow$ if $T_2 \ll T_1$ but are of physical interest even if the strong inequality does not hold. There are again two thermodynamic transitions associated with Bose condensation in the two spin states. A magnetization perpendicular to the stabilizing field appears below $T_{\lambda b}$ with a magnitude determined thermodynamically by the number density, n , S_z , and the temperature.

The changes in the thermodynamic functions due to H_1 can be calculated perturbatively. For the free energy per volume for $T < T_{\lambda b}$ we find

$$\delta F(N_a, N_b, T) = -\frac{\epsilon^2}{2} J [S_z^2 + n S_z + 2n^{\>}(T) S_z - \frac{1}{4} n^2] \quad (3)$$

where $n^{\>}(T)$ is the number of uncondensed particles

per volume in an ideal Bose gas below its transition temperature, and J is the average of V_E over a pair-distribution function determined from V_T .⁹ For low densities, $n \leq 10^{19}/\text{cm}^3$, $J \approx 4.2 \times 10^{-21} \text{ K cm}^3$ and is positive.⁹ The chemical potential difference,

$$\mu_b - \mu_a = H_z + \epsilon^2 J [S_z + \frac{1}{2} n + n^{\>}(T)] \quad (4)$$

sets the rotation rate of $\bar{\sigma}_1$.

Equations (2) and (3) imply immediately that the uniform state with $\bar{\sigma}_1 \neq 0$ ($T < T_{\lambda b}$) is thermodynamically unstable when $J > 0$ (antiferromagnetism) even though the energy gain per particle is 10^{-6} times smaller than $k_B T_{\lambda b}$. The two hyperfine states will move to separate regions of the container, in practice determined by inhomogeneities in H_z , until $\bar{\sigma}_1$ is reduced to zero everywhere, i.e., locally only one spin component is superfluid. Whereupon, F_0 in (2) becomes a function of N_a and N_b separately, and the usual entropy argument implies a termination of the phase separation.¹⁰

The remarkable "softness" of the $\sigma_1 \neq 0$ state can be understood following the example of a Heisenberg ferromagnet used above if a small anisotropy, DS_z^2 , is included. When $D < 0$ and the thermal value of $\langle \bar{S} \rangle$ exceeds the imposed value, domains will form with the magnetization either parallel or antiparallel to \hat{z} . Conversely, the opposite sign of D will stabilize the phase with $\sigma_1 \neq 0$.

If we restrict ourselves to times less than the phase-separation time, T_{ps} , but longer than T_2 or assume $J < 0$, the hydrodynamics of $H \downarrow$ for $T < T_{\lambda b}$ is correctly described by the theory of Andreev and Bashkin.¹¹ Since $H_1 \ll H_0$ and F_0 in (2) is only a function of the total density, the first- and second-sound modes are identical to those of a one-component superfluid of density n . The third propagating mode of Ref. 11 involves the concentration. Its velocity is complex if $J > 0$ with a magnitude set by $\epsilon^2 J n / m$.

In an ordinary fluid in a regime where the relative concentration moves with a diffusion constant D , one would estimate $T_{ps} \sim k_B T / (D \epsilon^2 J n) \sim 10^8$ sec for a 1-cm³ container. The simplest possible theory for $H \downarrow$ predicts that superfluid velocities of order $(\epsilon^2 J n / m)^{1/2} \sim 1$ cm/sec will arise within both spin components below $T_{\lambda b}$, thereby placing a lower limit of 10 sec on T_{ps} . The actual time could be far longer if $(\epsilon^2 J n / m)^{1/2}$ exceeded the relevant critical velocity.

The linearized hydrodynamics of Refs. 8 and 11 look very different, since in the former the broken symmetry is described by an overall phase and a direction, while in the latter, the order parameter is a pair of complex numbers with phases ϕ_a, ϕ_b . Actually, if we assume $J < 0$ or $T_2 \ll T_{ps}$ the two theories join smoothly. For frequencies less than T_2^{-1} , the spin-wave mode in the plane of H_z and $\langle \bar{S} \rangle$ is damped by T_2 while the remaining spin-wave mode and phase of Ref. 8 can be rewritten in terms of ϕ_a and ϕ_b .

The bulk relaxation times T_1 and T_2 have been calculated at temperatures below $\hbar^2/(2m\sigma^2) \sim 2$ K where σ is the radius of a hard-core approximation to V_1 . Conventional perturbation theory was done about a state in which the populations of a and b are in equilibrium so there is only one transition at $T = T_\lambda$ where "a" Bose condenses.¹² We assume $H_z/k_B T_\lambda \gg 1$, i.e., $n \leq 10^{18}$, and define a thermal wavelength, $\lambda = h/(2\pi mk_B T)^{1/2}$; and a magnetic moment, $\mu_\perp = \mu_p + \epsilon\mu_e$ where μ_p and μ_e are magnetic moments of the proton and electron.

At high temperatures,¹³

$$T_1^{-1} (\beta H_z \ll 1) = \frac{64}{15} \pi^{1/2} \mu_e^2 \mu_p^2 m^{3/2} n / (\hbar^4 \beta^{1/2}) \quad (5a)$$

while for all temperatures $\beta H_z \gg 1$, including $T \leq T_\lambda$,

$$T_1^{-1} (\beta H_z \gg 1) = \frac{16}{15} \pi \mu_e^2 \mu_p^2 m^{3/2} n H_z^{1/2} / \hbar^4 \quad (5b)$$

The dipolar contribution to T_2^{-1} equals T_1^{-1} both above and below T_λ to within numerical factors of order unity. In high fields, the exchange contribution to T_2 from (1b) was no larger than the dipolar term. High momentum, nonhydrodynamic degrees of freedom were found to dominate the relaxation at all temperatures, except to a limited extent in T_2 for $T \ll T_\lambda$.

One may also ask about possible collective contributions to the spin relaxation at low temperatures. The only such process we have found is speculative and would shorten only T_2 when a sample of H \downarrow was quenched to below $T_{\lambda b}$. Imagine the system to consist of clusters of uniform $\langle \vec{S} \rangle$ that move diffusively. A classical calculation then demonstrates that T_1 is set by the smallest clusters and T_2 by the largest.

The possibility of phase separation was overlooked in Ref. 4 so that even if T_1 were infinite and domains were ignored, states with $\sigma_\perp \neq 0$ only exist for a finite time. For similar reasons, the absence of inhomogeneous broadening would only be readily visible in a system with $J < 0$ since nonuniformities in the external field will pull the two hyperfine states apart below $T_{\lambda b}$ if $J \geq 0$. Thus even the model problem defined by setting H_1 and H_d to zero will phase separate in an inhomogeneous field below $T_{\lambda b}$ until the lower density spin component is no longer superfluid.

There are at least two classes of experiments that are potentially sensitive to Bose condensation in both hyperfine states or a local $\vec{\sigma}_\perp \neq 0$. The first requires T_2 significantly less than both T_1 and T_{ps} . Then, the relaxation of σ_\perp following a perpendicular pulse

would be different above and below $T_{\lambda b}$ since below it would relax to zero on the scale of T_{ps} or T_1 . Alternatively, one could imagine compressing a hydrogen sample until $T_{\lambda b}$ was raised above the ambient temperature. If a small rotating field were then applied the individual domains would be pulled into alignment on a time of order T_2 .

A second class of experiments would actually look for supercurrent assisted phase separation with spin echos. Assume the Zeeman energy in (1a) has the form

$$H_z(r) = H_z^0 + \delta H_z z^2 / \lambda_1^2$$

and that the sample occupies a region $|z| \leq \lambda_2 \ll \lambda_1$. In the normal phase, a spin echo will decay at a rate of order $[D \lambda_2^2 \delta H_z / (\lambda_1^4 h^2)]^{1/3}$, where $D \sim 10^{-2}$ cm²/sec is a diffusion constant and h is Planck's constant.¹⁴ Below $T_{\lambda b}$, there will be an additional contribution to the rate of order $(\delta H_z / m \lambda_1^2)^{1/2}$ which under favorable circumstances could exceed the diffusional term.

We wish to emphasize that the theoretical uncertainties in our estimates of the relative relaxation rates pertinent to either of the above experiments is sufficiently large as to be consistent with no effect at all below $T_{\lambda b}$. Perhaps in an appropriately designed cell the phase separation below $T_{\lambda b}$ could be followed on a macroscopic scale and its kinetics determined.

We have limited ourselves to a synopsis of what are perhaps the most unusual yet least accessible properties of the H \downarrow system; those associated with Bose condensation in both hyperfine states. Recombination has been totally ignored^{6,15} as well as properties that are sensitive to only a single condensate such as changes in the first-sound velocity or in the local density.¹⁶ Finally, much of our discussion could be repeated for adsorbed atomic hydrogen films if the conditions for Bose condensation prove easier to attain in two dimensions rather than three.¹⁷ In addition, a magnetic Bose fluid analogous to H \downarrow may yet be produced in an excitonic system.¹⁸

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¹L. H. Nosanow, J. Phys. (Paris) **41**, C7-1 (1980).

²A. J. Berlinsky, in Proceedings of the 26th Conference on Magnetism and Magnetic Materials, October 1980 [J. Appl. Phys. (in press)].

³I. F. Silvera and J. T. M. Walraven, in Proceedings of the 26th Conference on Magnetism and Magnetic Materials, October 1980 [J. Appl. Phys. (in press)]; and Phys. Rev. Lett. **44**, 164 (1980); R. W. Cline, J. J. Greytak, D.

- Kleppner, and D. A. Smith, *J. Phys. (Paris)* **40**, C7-15 (1980).
- ⁴E. D. Siggia and A. E. Ruckenstein, *Phys. Rev. Lett.* **44**, 1423 (1980); *J. Phys. (Paris)* **40**, C7-15 (1980).
- ⁵W. N. Hardy *et al.*, *Phys. Rev. Lett.* **45**, 453 (1980).
- ⁶B. W. Statt and A. J. Berlinsky, *Phys. Rev. Lett.* **45**, 2105 (1980).
- ⁷A. J. Berlinsky, *Phys. Rev. Lett.* **39**, 359 (1977).
- ⁸B. I. Halperin, *Phys. Rev. B* **11**, 178 (1975).
- ⁹L. J. Lantto and R. M. Nieminen, *J. Low Temp. Phys.* **37**, 1 (1979).
- ¹⁰M. D. Miller, *Phys. Rev. B* **18**, 4730 (1978). Miller finds phase separation only for bosons of unequal mass at zero temperature.
- ¹¹A. F. Andreev and F. P. Bashkin, *Zh. Eksp. Teor. Fiz.* **69**, 319 (1975) [*Sov. Phys. JETP* **42**, 164 (1976)].
- ¹²W. J. Mullin, *Phys. Rev. Lett.* **44**, 1420 (1980).
- ¹³The relaxation time, T_1 , in (5a) is one-half the value given in Ref. 6.
- ¹⁴A. Abragam, *Principles of Nuclear Magnetism* (Oxford University Press, London, 1961).
- ¹⁵M. Morrow, R. Jochemsen, A. J. Berlinsky, and W. N. Hardy, *Phys. Rev. Lett.* **46**, 195 (1981); A. P. M. Matthey, J. T. M. Walraven, and I. F. Silvera (unpublished).
- ¹⁶J. T. M. Walraven and I. F. Silvera, *Phys. Rev. Lett.* **44**, 168 (1980).
- ¹⁷I. F. Silvera and V. V. Goldman, *Phys. Rev. Lett.* **45**, 915 (1980); D. O. Edwards and I. B. Mantz, *J. Phys. (Paris)* **40**, C7-257 (1980).
- ¹⁸D. Hulin, A. Mysyrowicz, and C. Guillaume, *Phys. Rev. Lett.* **45**, 1970 (1980).