Explosive Recombination of Compressed Spin-Polarized Hydrogen

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(Received 29 October 1985)

Spin-polarized atomic hydrogen gas has been compressed to densities $> 10^{18}/\mathrm{cm}^3$ with liquid ^{4}He at temperatures from 0.24 to 0.67 K in magnetic fields up to 7.5 T. We find that the critical pressure, at which recombination ultimately becomes a thermally triggered explosion, goes through a maximum as a function of temperature. The explosion pressure increases with decreasing sample size and with increasing field and nuclear polarization.

PACS numbers: 67.40.Fd, 67.40.Kh, 68.10.Jy, 82.40.−g

In quest of observation of Bose-Einstein condensation (BEC) in spin-polarized atomic hydrogen (H↓), samples of this gas have been compressed to densities $n > 10^{18}$ cm^{-3} with a solid piston[1] or by flooding the sample cell with liquid ^{4}He,[2,3] and thereby decreasing the sample volume. Unfortunately, at high densities electronic dipolar interaction during a collision of three H↓ atoms becomes the dominant mechanism,[4] causing atoms even in the electron- and nuclear-polarized hyperfine state $b = |\downarrow \Downarrow\rangle$ to recombine into H_2 molecules both in the bulk gas and in the H↓ overlayer adsorbed on surfaces confining the sample. The third H↓ atom taking part in a three-body process can acquire enough of the recombination energy to become promoted to the electron-spin–reversed state $c = |\uparrow \Downarrow\rangle + \epsilon |\downarrow \Uparrow\rangle$ (where $\epsilon \simeq 0.025/B$, and B is the magnetic field in teslas) by the dipolar interaction. It will usually recombine with a fourth atom in a subsequent encounter before having a chance to relax back to the b state. The three-body recombination mediated by exchange interaction[4] cannot be ignored if there are atoms in the electronically mixed ground state $a = |\downarrow \Uparrow\rangle - \epsilon |\uparrow \Downarrow\rangle$. The presence of a atoms is due to the initial a-b mixture from the dissociator and to the nuclear $b \to a$ relaxation. At low densities the a-b and a-a pair collisions are the main cause of recombination. Their rates are, however, larger than the $b \to a$ relaxation rate and thus the nuclear polarization defined as $M = (n_a - n_b)/n$ evolves towards the value of -1. Yet at higher temperatures the a population will remain relatively large. At high temperatures and low fields also the $b \to c$ relaxation, the rate of which varies as $\exp(-2\mu B/k_B T)$, enhances recombination.

An exothermic reaction can be accelerated to terminate in an explosion if the increased heat release drives a corresponding temperature increase as a result of inadequate heat transfer to the surroundings. Sprik, Walraven, and Silvera[2] observed the recombination of compressed H↓ sometimes to proceed explosively if the ambient temperature of the sample was $T_0 \gtrsim 0.6$ K. This behavior was attributed[2,5] to the exponential temperature dependence of the $b \to c$ relaxation rate. We have reported[3] on the explosive recombination of H↓ bubbles even at 0.37 K. Here we describe a systematic study of compression sweeps at temperatures $T_0 = 0.24$–0.67 K and in magnetic fields $B = 4.5$–7.5 T. We find that a H↓ bubble, when compressed with superfluid ^{4}He, ends in an explosion which occurs at a maximum possible pressure P_c, determined by the recombination rate and the heat transfer. We anticipate that in the realm of the presently available experimental parameters explosions will frustrate attempts to attain conditions were H↓ undergoes BEC.

Our cryogenic system as well as the H↓ accumulation and compression techniques have been described in more detail elsewhere.[3,6] The compression is accomplished by flushing superfluid ^{4}He into the sample cell (SC) by means of a bellows system. The rising ^{4}He level first closes off the H inlet to the SC and then compresses the sample such that it is ultimately confined as a H↓ bubble to a flat cylindrical analyzing space (AS) in the top part of the SC. Two different copper SC's have been used in this work. In the larger one[3] the AS is 0.87 mm high and 20 mm in diameter while in the smaller SC these dimensions are respectively 0.22 and 2.0 mm. For improved thermal contact a silver sinter of 800 cm^2 is located at the entrance to the AS of the smaller SC. The AS's are confined between two gold-plated Kapton foils which provide the electrodes of a capacitance gauge for monitoring the volume V of the H↓ bubble via the amount of displaced ^{4}He. The resolution of the volume gauge limits the detectable sample size to $\gtrsim 0.01$ mm^3 in the large SC and to $\gtrsim 0.001$ mm^3 in the small SC. The ^{4}He level in the SC is determined from the amount of ^{4}He inside a coaxial capacitor which forms part of a tunnel-diode oscillator circuit. During the final phase of the compression it reads the hydrostatic pressure head P_h above the AS. The ambient temperature T_0 is measured with a calibrated carbon-film bolometer located on the outward-facing surface of the upper Kapton foil. The bolometer is capable of recording a short-lived transient heat pulse released in an explosive recombination of $\sim 10^{12}$ or more H↓ atoms.[7]

The H↓ sample is first compressed after loading of the SC to a density of 10^{13}–10^{15} cm^{-3}. Subsequently the SC can be recharged up to five times with the gas left over in the fill line from previous charging.[3] Usually the total low-density decay time does not exceed 20 min. The compression device is fully controllable from the outside and in this work P_h is increased at constant rates of 0.3–7 Pa/s once the sample is inside the AS. There the H↓ bubble first takes the shape of a disk bounded by the top and bottom Kapton foil. With further pressurization it shrinks in its horizontal diameter until it becomes small enough to attain a near-spherical shape and to be buoyed against the upper foil. The radii of curvature of the bubble now approach the uniform value $R=(3V/4\pi)^{1/3}$. The total pressure acting on the bubble is $P=P_h+P_s$, where $P_s=2\sigma/R$ is the surface-tension contribution with $\sigma=3.78\times10^{-4}$ N/m. As realized by Sprik, Walraven, and Silvera[2] P_h has to be corrected for the upward force which the magnetic field gradients exert on the diamagnetic ^{4}He column. For the highest critical hydrostatic head observed here, 39.6 mm ^{4}He, this correction was 5.5 Pa, but usually it was less than 1 Pa.

In the following we shall focus on the onset of the explosive recombination The general properties are illustrated in Fig. 1 where the critical pressure P_c is shown as a function of the bolometer temperature T_0 for different fields and H↓ bubble volumes at the onset of the explosion. Only nearly spherical bubbles with dimensions equal to or less than the height of the AS have been included in the graph. Their walls are assumed to reside uniformly at T_0. However, the apparent leveling of P_c at $T_0 \lesssim 0.35$ K can be understood on the basis of the increasing thermal resistance between the bolometer and the bubble, implying that our measured T_0 becomes increasingly lower than the actual surface temperature.[8] The H↓/^{4}He Kaptiza resistance is estimated to cause a temperature difference of about 0.09 K between the liquid surface and the gas in a $V_c=10^{-3}$-mm^3 bubble at $P_c=15$ Pa, $T_0=0.35$ K, and $B=7.5$ T if a value of 0.35 is accepted for the energy accomodation coefficient.[1,9]

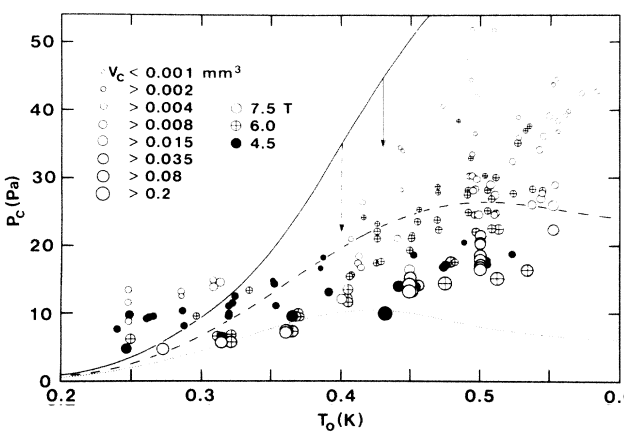

FIG. 1. Onset pressure P_c for explosive recombination of spherical H↓ samples as a function of the ambient temperature T_0. Different circle diameters correspond to different critical volumes V_c. For samples smaller than the volume detection limit the surface-tension contribution to P_c at this limit has been used. Open circles apply for $B=7.5$ T, crossed circles for 6.0 T, and closed circles for 4.5 T. The curves represent calculated pressures at maximum average density $\langle n \rangle_c$ of pure-b-state samples for $V_c=0.001$ mm^3 in 7.5 T (solid curve), 0.015 mm^3 in 6.0 T (dashed curve), and 0.1 mm^3 in 4.5 T (dotted curve). The right vertical solid arrow shows the effect on P_c of reduction of the nuclear polarization from $M=-1$ to $M=-0.85$. The left dashed arrow illustrates the influence of possible fluctuations triggering the explosion at $0.9\langle n \rangle_c$.

Figure 1 shows that the stability of the H↓ bubble improves with increasing polarizing field and decreasing sample volume. Both features grow more prominent when one moves from the surface-recombination–dominated regime at low temperatures to higher temperatures where recombination in the gas phase prevails and the strongly field-dependent electronic depolarization sets in. In this regime the poor thermal conductivity of the viscous H↓ gas[10] impairs transfer of the recombination heat from the bulk to the walls, which accounts for the pronounced dependence on the bubble diameter. In contrast to the clear T_0, V_c, and B dependences of P_c the compression rate is not an important consideration; i.e., the H↓ bubble transforms continuously from one thermal steady-state distribution to the next during the compression sweep. This experimental conclusion becomes evident upon considering the pressurization rate in the context of other time scales. The particle displacement due to the pressurization is at most of order $\dot{R}\, dn/dR \lesssim 10^{13}$ cm^{-3} at critical densities $n_c \gtrsim 10^{18}$ cm^{-3}. In our smallest bubbles with a diameter $2R \sim 100$ μm the mean free path is of order $\lambda \sim 5$ μm, resulting in a collision interval of $\tau_c \simeq \lambda/\bar{v} \sim 60$ ns, while the mean diffusion time between wall collisions is $\tau_D \simeq 6R_c^2/\bar{v}\lambda \sim 60$ μs. Experimentally the explosion is observed to take place in less than 30 ms, limited by the rise time of the volume gauge.

Following Kagan, Shlyapnikov, and Vartanyantz[5] we have employed a simple approach to model explosions in the H↓ bubbles. We assume that the entire recombination heating Q is distributed homogeneously over the bubble volume.[8] Because $R_c >> \lambda$, the heat flux in the gas can be treated as normal heat conduction in a viscous medium, yielding a parabolic temperature profile $T(r)=T_0+(q/6K)(R^2-r^2)$ for a sphere. Here $q=Q/V_c=(D/2)\,dn/dt$, where $D=4.5$ eV and the recombination rate (dn/dt) has been obtained by using the most recent experimental or calculated rate coefficients[1,2,4,11] for the various three- and two-body volume and surface decay processes. As calculated by Lhuillier[10] the thermal conductivity of H↓ is almost constant, $K \simeq 3.6$ mW/K·m, in the present range of temperatures. For constant V_c, T_0, B, and M we integrate the average gas-phase density $\langle n \rangle = N/V_c$ at different pressures, which are assumed to be uniform throughout the volume. Because volume recombination depends on $T(r)$, q and $T(r)$ have to be iterated self-consistently. The critical pressure P_c for the violation of heat balance is identified[5] as the point where $\langle n \rangle$ obtains its maximum as a function of temperature, i.e., $d\langle n \rangle / dT(0) \to 0$. Pressures higher than P_c represent a physically unjustifiable solution of increasing $T(0)$ with decreasing density. Because $dT(0)/d\langle n \rangle$ becomes very large for $P \lesssim P_c$, small density fluctuations may trigger the H↓ system from stable behavior to thermal runaway earlier than expected (cf. Fig. 1).

Figure 2 illustrates the observed volume dependence of P_c which at different T_0 appears to be roughly of the form $P_c \propto V^{-1/\gamma}$, where the exponent γ is determined by the prevailing recombination mechanism. If the three-body surface recombination[12] is the only heat-generating process, the model described above predicts

$$P_c = \left(\frac{2T_R K}{DL_s R_c} \right)^{1/3} \frac{k_B T_R}{\Lambda_R} e^{-E_a/k_B T_R}, \quad (1)$$

where L_s is the intrinsic rate coefficient,[1] $T_R = T(R_c)$, $\Lambda_R \propto T_R^{-1/2}$ the thermal wavelength of a H atom at the ^{4}He surface, and E_a the adsorption energy $\simeq 1.0$ K.[1,11] According to Eq. (1) $\gamma = 9$, while for the three-body volume recombination the model gives $\gamma = 4.5$. The experimental value in Fig. 2 ranges from $\gamma = 8.1$ at $T_0 = 0.325$ K to $\gamma = 5.8$ at $T_0 = 0.51$ K. Clearly both surface and volume processes are thus of importance in this temperature interval, although towards lower temperatures the surface recombination is rapidly growing dominant. This feature is further demonstrated by Fig. 3 where $\ln P_c$ has been plotted vs $1/T_0$ for constant values of V_c and B. The average slope of the fitted solid lines is -1.08 K, i.e., roughly equal to E_a in the low-temperature regime, while at higher T_0 the data deviate from the linear dependence.

Above 0.5 K the atomic-hydrogen bubbles again exhibit a reduced stability towards thermal explosion, in fact, often to such a degree that they explode well before reaching the spherical shape. This is, in particular, the case for large samples which have been compressed immediately after their accumulation such that their nuclear polarization M is initially close to zero. As can be calculated from the rate equations controlling the recombination decay,[1,2,11] M evolves more slowly during compressions at high temperatures. Thus, in addition to the three-body dipolar and the b-c depolarization processes in the gas, recombinations involving a atoms now add to the heating rate. Other reasons, which contribute to the reduction of P_c, are the finite density of ^{4}He-vapor atoms and the presence of excited H_2 molecules. Also, above 0.5 K, P_c is found to decrease with increasing bubble volume and with decreasing polarizing field.

In summary, this work shows that when the H↓ density is increased by compressing the sample inside superfluid ^{4}He the gas will ultimately undergo an explosive recombination at a reproducible pressure P_c which goes through a maximum as a function of the ambient temperature T_0. Above this maximum, P_c decreases as a result of the three-body recombination and electronic depolarization in the gas phase. In this region the incompleteness of nuclear polarization also becomes an important limitation on P_c. The highest H↓ pressure, $P_c > 63$ Pa, was achieved at $T_0 = 0.54$ K and $B = 7.5$ T for a bubble with a radius clearly smaller than the resolution $R = 60\ \mu$m of our volume gauge. Assuming $T_R = T_0$ we obtain $n(R_c) > 8.5 \times 10^{18}$ cm^{-3} for the maximum density, $\langle n \rangle_c \gtrsim 5.7 \times 10^{18}$ cm^{-3} for the average density, and $T(0) \approx 1.2$ K for the center temperature. These densities are still a factor of at least 20 short of the critical density required for BEC at 0.54 K.

Towards lower temperatures P_c is also exponentially

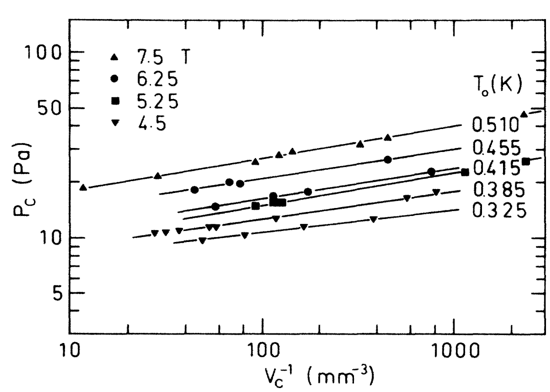

FIG. 2. P_c vs the inverse of the spherical H↓ bubble volume V_c at different ambient temperatures T_0. The lines are fits to the data with $P_c \propto V_c^{-1/\gamma}$ dependence.

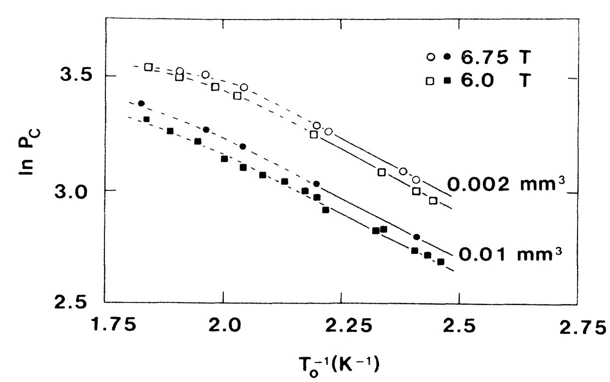

FIG. 3. $\ln P_c$ vs $1/T_0$ for two sets of values for V_c and B. The dominant influence of three-body surface recombination at low T_0 is displayed.

limited but now by the three-body recombination on the surfaces [cf. Eq. (1)]; i.e., it drops more rapidly than the BEC pressure $\propto T_0^{5/2}$. The relatively slow variations of the recombination rates with magnetic field[1] and of P_c with volume [cf. Eq. (1) and Fig. 2] are not expected to allow BEC densities to be obtained with the experimentally challenging parameters of $V_c = 10^{-6}$ mm^3 and $B = 15$ T and even in the absence of thermal boundary resistances. However, the surface densities required for the onset of a Kosterlitz-Thouless superfluid transition in the adsorbed H↓ monolayer on ^{4}He would be attained in $R = 6$ μm bubbles at $T_0 \lesssim 0.3$ K. Adding ^{3}He into the ^{4}He at $T_0 \lesssim 0.2$ K, where a ^{3}He surface coating is formed, would result in threefold reduction in E_a and would enhance P_c by a factor of about 20. Even this would not, however, be enough for achieving BEC. At higher temperatures the higher vapor pressure of ^{3}He leads to increasing volume recombination and lowers P_c as has been observed in these and earlier[2] experiments.

The financial support of the Academy of Finland, the Wihuri Foundation, and the Turku University Foundation is gratefully acknowledged.

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[7]The existence of bubbles with fewer atoms is sometimes revealed by the shape (but not the magnitude) of the volume-gauge signal during compression, but a possible explosion remains undetected.

[8]At $T_0 \lesssim 0.35$ K the bubbles seem to tolerate higher pressures than expected and this may be partly attributed to the fact that some fraction of the surface recombination heat is dumped directly into the ^{4}He bath.

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