

## EXPERIMENTAL STUDY OF SPIN ALIGNED ATOMIC HYDROGEN CONDENSED ON SURFACES

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Received 13 March 1978

A study is made of spin aligned atomic hydrogen ( $H\uparrow$ ) condensed on Ar and  $H_2$  surfaces in order to study the stability of H in a condensed phase at helium temperatures in a magnetic field. Results are consistent with a picture of rapid spin depolarization on the surface followed by desorption or recombination.

Atomic hydrogen (H) is predicted to have a number of spectacular properties in the low temperature condensed phase, however very little experimental information is available because of its instability toward recombination to the molecular state. At low densities ( $\rho^{-1} > 50 \text{ cm}^3/\text{mole}$ ) it has been speculated that it can be stabilized by imposing a large magnetic field to align the spins, forcing the atoms to interact in the nonbinding  $^3\Sigma_u^+$  potential. In this case one can predict Bose condensation of the spin aligned hydrogen ( $H\uparrow$ ) as a magnetic superfluid gas. For the past several years we have been developing a number of ideas for the production, stabilization and study of H [1]. In this letter we describe experiments in which H is produced by dissociation of  $H_2$  in a microwave discharge, formed into an atomic beam, spin polarized and deposited on a helium cooled substrate in a weak magnetic field.

The surface is used to localize the atoms in space enabling the build up of a useful density in a relatively short time. For certain types of surfaces (consisting of closed shell atoms or molecules), the adsorbed H atoms will remain neutral, the substrate potential serving to bind the atoms to the surface. In this case spin polarization of the atoms can be meaningful as the single atom properties are not expected to be severely modified by the surface. The H atom-atom interactions should be well described by the spin dependent potentials calculated by Kofos and Wolniewicz [2]. The potential corresponding to the electron spin singlet,  $^1\Sigma_g^+$  is binding and representative for unpolarized condensed H which should rapidly recombine to  $H_2$ .

The triplet  $^3\Sigma_u^+$  state is non-binding and is representative for interactions amongst  $H\uparrow$  atoms, and if the spins do not flip the condensate should be stabilized. The objective of the measurements described here was to investigate the use of spin polarization and small magnetic fields on the build-up of a surface condensate. The results are that no measurable condensate has been built up. The most likely cause is spin depolarization on the surface followed by desorption or rapid recombination. Knowledge of the phenomena studied are of interest for H-surface interactions and vital for the attempts being carried out in this and other laboratories to stabilize  $H\uparrow$  in a gas form, surrounded by surfaces.

Neutral H, possessing an electron spin  $S = \frac{1}{2}$  and nuclear spin  $I = \frac{1}{2}$  ( $F = I + S = 0, 1$ ), is a composite boson (as helium) however for reasons of stability, only  $H\uparrow$  is of interest. The possibility of superfluidity and the non-existence of a solid or liquid phase was first discussed by Hecht [3]; more recently there has been a newly born interest for calculations of the thermodynamic properties [4,1] and consideration of the elementary excitations [5]. The experimental work that has been done in this field can be roughly divided into matrix isolation and atomic beam work. In the former H is created in a discharge and condensed, along with a large percentage  $H_2$ , in a cold region, possibly in a large magnetic field [6] or is created at lattice sites in solid  $H_2$  by a radioactive decay scheme [7]. Small amounts of H are apparently stabilized by matrix isolation, however this type of experiment does not appear to enable the study of the weakly interacting H system

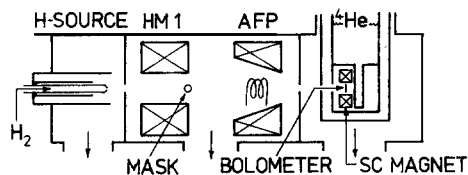


Fig. 1. Apparatus for polarization measurements.

as the matrix imposes dominating boundary conditions on the H atoms. The second type of experiments carried out have been with atomic beams [8,9]. Here *unpolarized* beams of purity  $\sim 80\text{--}90\%$  H ( $\text{H}_2$  impurity) are deposited on cold substrates ( $\sim 4$  K). The conclusions drawn are that only a small amount of the H condenses on the substrate and that this rapidly recombines, the resulting  $\text{H}_2$  molecules desorbing to carry off the large recombination energy (4.5 eV). Here matrix isolation can also occur if the  $\text{H}_2$  content of the beam is high and the H is only a small fraction of the adsorbed particles, or if the surface has deep trapping sites for the H atoms.

In our experiments we work with an ultra high purity room temperature H beam that can be placed in various states of spin polarization. The H is deposited on a cold (1.8–4.2 K) non-magnetic surface of argon or other closed shell atomic or molecular species. Such surfaces can weakly bind the H atoms by virtue of the van der Waals attraction; if the potential has weak variations in the surface plane, H atoms will not be localized. Thus, in a simplified picture we envisage adsorbed H to behave as a quasi two-dimensional gas interacting according to the Kofos–Wolniewicz potentials. These systems have the nice property that recombination of H does not lead to  $\text{H}_2$  impurities on the surface as the latter is expected to desorb due to the large recombination energy [8,10]. The main experimental difficulty is the extremely small amount of sample (a small fraction of a monolayer) that one has to study. As a result we do not have a direct method to measure the H coverage on the surface. Instead we measure the heat flux to the surface due to the H beam by means of a silicon bolometer ( $5 \times 5 \text{ mm}^2$ ), sensitive to an H atom flux of  $\sim 10^8$  per second. The bolometer can be precoated in situ with argon, etc., to provide a clean surface.

A schematic diagram of the experiment is shown in fig. 1. The H beam is formed by means of a microwave discharge [11] yielding a degree of dissociation of  $\sim 85\%$ .

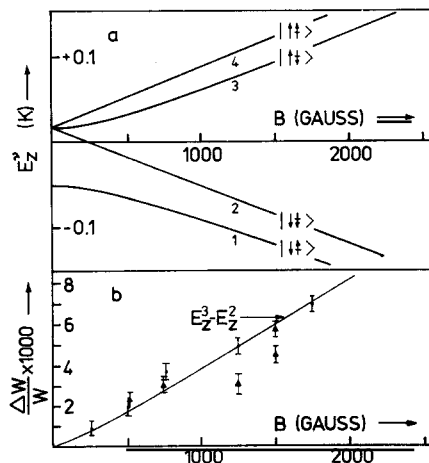


Fig. 2. (a) Hyperfine level diagram for H. The electronic (†) and nuclear (‡) high field spin states are shown. (b) Points: ac bolometer signal as function of field for Ar surface; line: fit of hyperfine splitting to data.

It is then polarized by means of a hexapole magnet (HM1) which focuses atoms in hyperfine states 3 and 4 (shown in fig. 2a) on the bolometer, while atoms in states 1 and 2 are bent out of the beam. The polarization is defined as  $P = (p\uparrow - p\downarrow)/(p\uparrow + p\downarrow)$  where  $p\uparrow$  and  $p\downarrow$  are the probabilities of finding an electron spin up or down. By masking off the axis of HM1, most of the molecules can be removed to achieve a beam purity of 0.9994, which to our knowledge exceeds previous H beam purities by about two orders of magnitude. A flux of  $\sim 10^{14}$  atoms/cm<sup>2</sup>/s is incident on the substrate. The mean velocity of the H atoms was calculated [12] to be  $2.7 \times 10^5$  cm/s with a 10% spread.

The beam can be depolarized by inverting the populations of states 3 and 2 by an adiabatic fast passage [13] (AFP) scheme. This is accomplished by passing the beam through an rf loop in a magnetic field gradient. We have measured  $\Delta P \approx 0.85$ . By modulating the rf power on and off,  $P$  can be varied without affecting the beam intensity. The bolometer is inside the bore of a small superconducting (SC) magnet which has a maximum operational field of  $\sim 2.5$  kG. The magnet can be used to place the atoms in the high field limit with respect to the hyperfine field ( $B \gg 507$  G) so that states 1 and 3 are almost pure electron spin down and up states. Schutte et al. [8] have shown that changes in surface composition can be studied by

measuring the power dissipated in a bolometer by a beam. We have used this technique to study the effect of spin polarization on the condensation properties of H on Ar and H on H<sub>2</sub> surfaces. For H on Ar we measured a small (ac) signal due to modulating the polarization of the beam, on top of a large (dc) signal. This ac signal corresponded to an increased heating of the bolometer upon polarizing the beam and was about linearly dependent on the SC magnetic field as shown in fig. 2b. The signal increased to  $\approx 0.8\%$  of the dc signal at  $B \approx 2$  kG. This ac signal vanished for H on H<sub>2</sub> surfaces. The signal was independent of modulation frequency in the range of  $5-10^3$  Hz as limited by the bolometer response. The observations can be understood if we assume that we are detecting a difference in Zeeman energy between the two polarization states of the beam. This small effect is intrinsic to the method, shows the observed field dependence and can account for an ac heating of  $\sim 10^{-11}$  W, roughly the order of the signal that was observed for an SC field  $B \approx 2$  kG and an estimated collimated flux  $\Phi_0 \approx 6 \times 10^{12}$  atoms/s. Using this model the bolometer signal is proportional to  $W = \sum W^\nu$  where  $W^\nu = W_s^\nu + W_{1-s}^\nu$  denotes the thermal flux due to particles in hyperfine state  $\nu$ :

$$W^\nu = s\Phi_0^\nu \{ \alpha_s [E_k + E_k^\nu(B)] + E_a^H + E_z^\nu(B) \} \\ + \alpha_r E_r - \frac{1}{2} \alpha_a E_a^{H_2} \\ + (1-s)\Phi_0^\nu \{ \alpha_{1-s} [E_k + E_k^\nu(B)] + \alpha_z E_z^\nu(B) \}. \quad (1)$$

Here  $s\Phi_0^\nu$  is the fraction of the incident flux  $\Phi_0^\nu$  of atoms in state  $\nu$  that adsorb and recombine or remain as H atoms; the second term represents the fraction  $(1-s)\Phi_0^\nu$  that have inelastic collisions but do not adsorb.  $E_k$  is the kinetic energy of a beam particle for  $B=0$  and  $E_k^\nu(B)$  is the incremental change of  $E_k$  for a particle in state  $\nu$  accelerated into the field  $B$ ;  $E_z^\nu(B)$  is the corresponding Zeeman energy. Note that

$$E_k^\nu(B) = E_z^\nu(0) - E_z^\nu(B). \quad (2)$$

$E_r$  is the recombination energy and  $E_a^H$  and  $E_a^{H_2}$  are adsorption energies, and the coefficients  $\alpha$  are fractional amounts of the corresponding energies dissipated in the surface. Quantities not indexed by  $\nu$  are assumed to have a negligible dependence on the polarization. Eq. (1) is valid for  $kT \gg \Delta E_z$ , the hyperfine splitting. One might expect  $W_s$  to be dominated by  $E_r \approx 52000$  K, but as has been shown by Schutte et al. and reconfirmed by us [1],  $E_r$  is not dissipated in the surface but carried

away by desorbing H<sub>2</sub>; thus  $\alpha_r \lesssim 0.01$ . For an H<sub>2</sub> surface, the H beam atoms are easily accommodated due to the similar masses. Roughly, we expect  $s \approx \alpha_s \approx 1$ , resulting in  $W \approx W_s$ . When modulating between states 3,4 and 2,4, the modulation signal is  $\Delta W = W^3 - W^2$ . In this case  $\Delta W = 0$  due to eq. (2), i.e. an atom that comes into thermal equilibrium with the surface cannot contribute to  $\Delta W$ . This apparently explains the experimental result for H<sub>2</sub> surfaces. For H on Ar we are in the other limit with  $W \approx W_{1-s}$ . In order to explain the signal we are led to assume that  $\alpha_z \approx 1$  and  $\alpha_{1-s} \ll 1$ . This means that nonadsorbing atoms have sufficient residence time on the surface to spin thermalize (depolarize for  $kT \gg \Delta E_z$ ), however insufficient time for translational accommodation. We then find

$$\Delta W = \frac{1}{2} (1-s) \Phi_0 (\alpha_z - \alpha_{1-s}) [E_z^3(B) - E_z^2(B)]. \quad (3)$$

A scaled form of eq. (3) is shown as the solid line in fig. 2b, comparing favorably with experiment. A difficulty that arises here is the scale. For Ar we have measured  $W/\Delta W \approx 125$  for  $B = 2$  kG. Using  $E_z^3 - E_z^2 \approx 0.24$  K we find a maximum value of  $E_1 \equiv W/\Phi_0 = 15$  K/atom. Schutte et al. measured  $E_1 \approx 1100$  K/atom for H on H<sub>2</sub>. We have also made absolute measurements of  $W$  under similar conditions (without HM1) and find  $E_1 \approx 2900$  K/atom. However, using HM1 we find<sup>#1</sup>  $E_1 \approx 370$  K/atom for H on H<sub>2</sub> and  $E_1 \approx 123$  K/atom for H on Ar. The main difference in using a hexapole is the change in velocity distribution from a broad velocity weighted maxwellian of 500–600 K to a very peaked distribution around 443 K. Thus the accommodation has a remarkably strong dependence on  $E_k$ . A discrepancy of about 8 exists for  $W$  determined from magnetic considerations and by absolute calibration. We note that contributions to  $\Delta W$  can also arise from  $W_s$  since  $\alpha_s$  can be less than one.

We thus visualize the process as one in which hydrogen atoms that impinge on the surface rapidly depolarize, possibly via magnetic dipole–dipole (with H or magnetic impurities) or hyperfine interactions as suggested by the calculations of Berlinsky et al. [14]. The atoms

<sup>#1</sup> The influence of HM1 on beam properties has been determined by phase space calculations (see Audoin et al. [12]). The intensity determination with HM1 in the beam must be considered as a reasonable estimate rather than an absolute measurement due to this calculational step.

that stick, rapidly recombine and eject from the surface as  $H_2$ . We estimate an upper bound on the coverage of  $10^{11}$  atoms/cm<sup>2</sup>, with atoms possibly trapped at surface sites.

Finally we point out that a condensate in states 3 and 4 cannot be stabilized by a magnetic field. The required conditions are that the atoms be in states 1 and 2 in the presence of a high field and at low translational temperatures [14]. A beam can be prepared in these states by AFP and deposited in an intense magnetic field, but it appears that achievement of the stability conditions as discussed in ref. [14] and study of  $H\uparrow$  would be easier in a three dimensional (3D) gas rather than on a surface. However, in a real system, if the atoms of a 3D gas are in equilibrium with the surface, then to achieve stability, both components must be stabilized. Thus an understanding of surface phenomena is vital.

We would like to thank H. Godfried, A.J. Berlinsky, G. Scoles and F. Tommasini for a number of stimulating interactions. The assistance of O. Höpfner in the construction of the apparatus has been of immeasurable value. We gratefully acknowledge the Stichting FOM for financial support.

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