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Optical Excitation of Atomic Hydrogen Bound to the Surface of Liquid Helium

A. P. Mosk, M. W. Reynolds, and T. W. Hijmans

Van der Waals–Zeeman Instituut, Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

J. T. M. Walraven

FOM Instituut voor Atoom en Molecuulfysica (AMOLF), Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

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We have optically detected hydrogen atoms adsorbed on the surface of liquid helium, a system relevant for the study of Bose degeneracy in two dimensions. The atoms are excited by 121.6 nm light and detected both in fluorescence and in absorption. The excitation spectrum shows a resonance that is much broader than that of a free atom, and shifted to lower frequencies. From the absorption signal we determine that we have reached a surface density corresponding to one atom per square de Broglie wavelength. We also use the hydrogen fluorescence to probe the thermal resistance between the free surface and the bulk of the liquid helium. [S0031-9007(98)07592-9]

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Spin-polarized atomic hydrogen adsorbed on the surface of liquid helium is a unique example of a two-dimensional (2D) atomic gas. This 2D gas is exceptionally stable which allows for the accumulation of large surface densities. In addition, the atoms are so weakly bound to the helium surface that it is possible to maintain an equilibrium between the adsorbed gas and the 3D gas of nonadsorbed H atoms. These properties make it a ready model system for investigating quantum degenerate behavior in two dimensions.

The goal of reaching the quantum degenerate regime in the 2D Bose gas of adsorbed H has been the subject of several studies (see, e.g., Refs. [1,2]). Recently, this goal has been achieved [3]. Crucially, however, in these experiments the 2D Bose gas itself has only been observed by indirect means. The surface density was inferred from recombination rates. The geometry required to approach degeneracy involves compressing the gas onto a small effective surface in a strongly inhomogeneous magnetic field, making magnetic resonance techniques (as used in, e.g., Ref. [4]) unsuitable. As a consequence, direct observation of the 2D gas is a nontrivial matter. In this paper we present the first measurements in which we have direct access to the compressed surface gas in such a geometry. For this purpose we use resonant optical spectroscopy of the adsorbed H atoms near the $1S-2P$ (Lyman- α) transition. The adsorbed atoms were anticipated to give rise to a surface specific fluorescence signal [5,6] but the precise frequency, line shape, and width of the resonance were not known in advance. In this Letter we show that the fluorescence spectrum can be used to determine the surface density under the conditions relevant for studies of 2D quantum degeneracy.

In our experiments we obtain a 2D degeneracy parameter $n_2\Lambda^2$ close to unity, where n_2 is the surface density and Λ is the thermal de Broglie wavelength. This means that we are at the edge of the quantum degenerate regime. Theory predicts a Kosterlitz-Thouless-type phase transition to

superfluidity at $n_2\Lambda^2 \approx 4$. The formation of a quasicondensate is predicted to occur close to this transition [5].

We use the adsorbed H atoms to probe the coupling between the excitations of the He surface (ripples) and those of the bulk liquid (phonons) [7]. This coupling determines the temperature of the surface in the presence of recombination heating. The surface density depends exponentially on temperature. As a consequence, the thermal resistance between the ripples (to which the H atoms couple strongly) and phonons turns out to be the dominant constraint on achievable phase-space densities. We find good agreement between our observations and theory describing ripplon-phonon coupling.

In our experiments we determine, in addition to n_2 , two other relevant thermodynamic quantities: the temperature T , and the density n_3 of atoms in the 3D gas above the surface. The 3D gas density fixes the chemical potential, thus giving access to the equation of state of the 2D Bose gas. Following Ref. [5] this equation of state can be written in the following slightly simplified form:

$$n_3\Lambda^3 = (1 - e^{-n_2\Lambda^2}) \exp\left(\frac{-E_b + (2 - \Theta)\tilde{U}n_2}{k_B T}\right). \quad (1)$$

Here $\Lambda \equiv (2\pi\hbar^2/mk_B T)^{1/2}$ with m the atomic mass, $E_b/k_B = 1.01$ K [8] is the binding energy of H on ^4He , and $\tilde{U}/k_B = 5 \times 10^{-15}$ K cm 2 [9] is the calculated effective interaction vertex between adsorbed atoms. The parameter Θ is a measure of the quasicondensate density; Θ is zero in the absence of a quasicondensate and becomes approximately 1 when $T \rightarrow 0$. Hence, Eq. (1) can be used as a telltale sign for a possible transition to a Bose condensed state.

Our cryogenic apparatus [10] is shown schematically in Fig. 1. The H is stabilized by a magnetic field of several tesla and a liquid helium film wetting all surfaces. The apparatus consists of a small cold region called the cell, in which the atoms adsorb on a meniscus of bulk liquid helium, and a warmer buffer volume, which acts as a

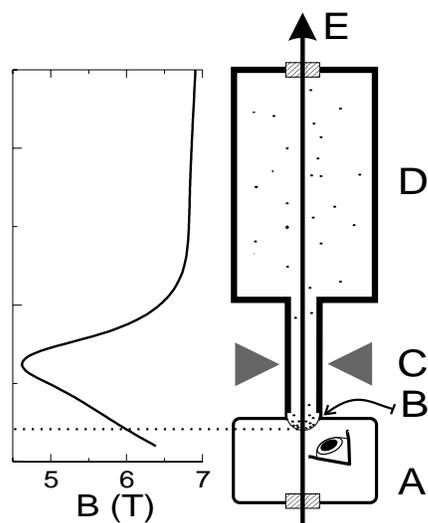


FIG. 1. Schematic drawing of the cryogenic apparatus showing (A) the He filled cell ($T = 0.08$ K, effective area 4 mm^2), (B) the He meniscus with the high density H, (C) the magnetic potential barrier (pinch), D the buffer ($T = 0.2\text{--}0.4$ K, effective volume 30 cm^3), (E) the path of the light beam. Plotted alongside is the magnitude of the magnetic field B on the axis of the system. The magnetic field is essentially vertical everywhere.

source of doubly (electron and nuclear spin) polarized H for the cell. The cell is cooled to 0.08 K, while the buffer temperature can be regulated between 0.15 and 0.4 K.

A superconducting magnet and iron pole pieces create a magnetic field profile as shown in Fig. 1. This provides a high field in the buffer, a strong field gradient in the cell and a deep dip in the field in between the buffer and the cell, which acts as a barrier for spin-down H atoms moving between the buffer and the cell.

A cryogenic dissociator in low field produces a flux of 10^{13} atoms per second into the buffer. These atoms are in the high field seeking a ($F = 0$) and b ($F = 1, m_F = -1$) hyperfine states. Importantly, although the cell is much smaller than the buffer, it determines the loss of atoms because it is at lower temperature. The two- and three-body decay rates are dominated by recombination on the surface. Hence, it follows from Eq. (1) that in the nondegenerate case they are proportional to $(n_2)^2 \propto \exp(2E_b/k_B T)$ and $(n_2)^3 \propto \exp(3E_b/k_B T)$, respectively.

The main two-particle recombination processes for adsorbed atoms are $a + a \rightarrow \text{H}_2$ and $a + b \rightarrow \text{H}_2$. The first leads to the polarization of the gas to the b state and the second leads just to loss of density. The process $b + b \rightarrow \text{H}_2$ is forbidden. Three-body processes become dominant over the $a + b$ recombination only after a substantial degree of nuclear polarization has been achieved. Once the nuclear spin polarization is sufficient, we heat the buffer volume so that the flux of atoms to the cell increases. The density in the cell is then built up by this forward flux, until it reaches a stationary state due to the backward flux (from the cell to the buffer) and

the three-particle recombination process $b + b + b \rightarrow \text{H}_2 + \text{H}$ of the adsorbed atoms in the cell. Because the cell is much colder than the buffer, the backward flux is inhibited and the density in the cell becomes much higher than that in the buffer.

We use narrowband vacuum ultraviolet (VUV) light to determine the densities n_2 and n_3 and the temperature T in the cell, as well as the density of the gas in the buffer. We produce this VUV radiation at 121.6 nm wavelength by frequency tripling of a pulsed UV laser source [11]. The VUV light traverses the cell and buffer on the axis of the system. A room temperature photomultiplier tube (PMT) detects the transmitted light. We can also record light-induced fluorescence (LIF) spectra by scanning the laser frequency. The scattered light is frequency downconverted to visible wavelengths in the cell and guided to a second PMT by means of an optical fiber bundle.

As the magnetic field is parallel to the direction of propagation of the light, only σ_+ ($\Delta m_J = 1$) and σ_- ($\Delta m_J = -1$) transitions can be excited. Moreover, as the field is high (>6 T) the spin-orbit coupling is weak, and the $1S_{1/2,-1/2} \rightarrow 2P_{3/2,1/2}$ transition is strongly suppressed and remains nonsaturated even when allowed lines are extremely optically thick. In Fig. 2 we show a spectrum of this line. The different magnetic fields in the cell and buffer split the line into two distinguishable components. The strongly inhomogeneous field in the cell gives rise to a temperature dependent Zeeman broadening (on the red wing of the cell line) which we fit against calculated spectra to determine the temperature of the 3D gas.

Our measurements of the spectrum of the adsorbed atoms, performed with σ_- polarization, show an ≈ 300 GHz broad line, which can be seen both in LIF and in transmission and which is shifted ≈ 300 GHz to

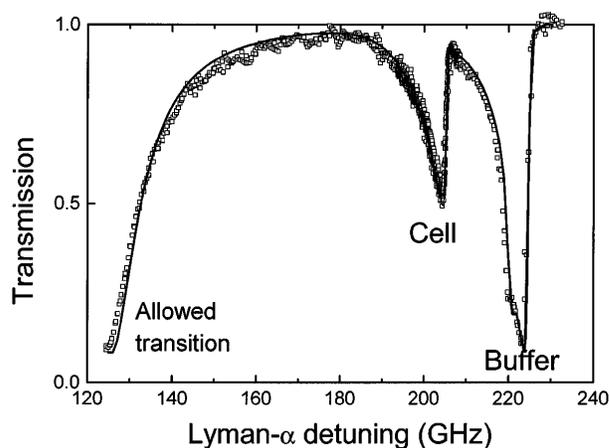


FIG. 2. 3D H transmission spectrum on the nearly forbidden $1S_{1/2,-1/2} \rightarrow 2P_{3/2,1/2}$ transition. Squares: experimental data; line: calculated spectrum for $T = 0.122$ K, $n_3 = 1.3 \times 10^{15} \text{ cm}^{-3}$, polarization of the light 93% σ_+ . The strong feature on the left is the far wing of the allowed $1S_{1/2,-1/2} \rightarrow 2P_{1/2,1/2}$ line.

the red of the allowed σ_- bulk line. A similar feature is observed with σ_+ light but it is obscured by a coincident 3D H line. The bottom panel in Fig. 3 shows the LIF signal of the σ_- surface line.

The observed line shift and broadening are very small compared to the detunings with respect to all other optical transitions in H and He. Therefore, in analogy with pressure broadening in a gas, we assume that the He-H interaction conserves the oscillator strength of the Lyman- α transition to a good approximation. This assumption, together with the measurement of the optical extinction spectrum of the surface atoms, enables us, in principle, to obtain an absolute calibration of n_2 . However, parts of the line are masked by the much stronger 3D H lines, or lie outside the scanning range of our light source. Integrating the visible part of the 2D H optical resonance therefore provides us with a lower bound to n_2 . We can also establish an upper bound to n_2 from the rate of recombination as follows: In the case of perfect nuclear spin polarization, n_2 can be inferred from the known rate of $b + b + b$ recombination [12]. If the polarization is imperfect, the recombination is necessarily faster for a given n_2 , and the inferred n_2 will be an upper bound. We find this upper bound to be only 25% higher than the lower bound determined optically.

Equation (1) suggests that quantum degeneracy will be reached if the flux from the buffer into the cell is simply increased at fixed cell temperature. This increases n_3 , and n_2 will adjust accordingly. Unfortunately, this picture is too simple because it neglects the temperature rise of the surface due to increased recombination. This temperature rise in turn reduces n_2 . To interpret our measurements we

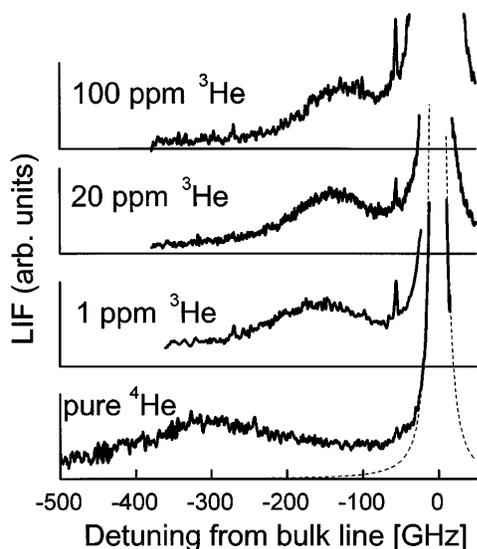


FIG. 3. LIF excitation spectrum of the adsorbed atoms at different concentrations of ^3He . Note that 100 ppm ^3He corresponds to well-saturated coverage of the surface. The dotted line is a Lorentzian fit to the bulk line. The narrow peak at -50 GHz is due to photoassociation of H atoms in the bulk.

introduce three temperatures: the temperature T_3 of the 3D gas, the temperature T_2 of the 2D gas, and the temperature T_{He} of the bulk helium. Two of these three temperatures are directly accessible experimentally: T_3 is determined spectroscopically (from the Zeeman broadened line shape) and T_{He} is measured with a resistance thermometer inside the liquid. In addition we are able to determine T_2 from our spectroscopic measurements of n_2 and n_3 . The temperature drop between the 3D H and the surface is small enough that solving Eq. (1) for T yields a good estimate of the surface temperature. For larger temperature drops, T_2 can be inferred from the more general equations in Ref. [5].

Figure 4 shows the three temperatures T_3 , T_2 , and T_{He} as functions of the recombination rate. This rate follows directly from the decay of the total number of atoms in the apparatus. It is clearly seen that the temperatures become different as the recombination rate increases. This can be understood quantitatively as follows.

In a three-body recombination event only a small fraction (less than 1.4% [2]) of the 4.5 eV binding energy of H_2 is deposited to the surface as heat; the rest is carried away as molecular internal energy. Subsequently, the excited molecule requires ≈ 150 wall collisions to release half its energy [12]. During its excited lifetime, the molecule may return to the helium meniscus and deposit part of its energy there. Assuming diffuse scattering of molecules from the walls, we calculate that in our cell the molecules deposit 3% of the recombination energy on the surface of the meniscus. This gives rise to a temperature drop between the surface and bulk of the liquid helium. The magnitude of the temperature drop we find here confirms that the ripples of the He surface are weakly coupled to the phonons in the superfluid

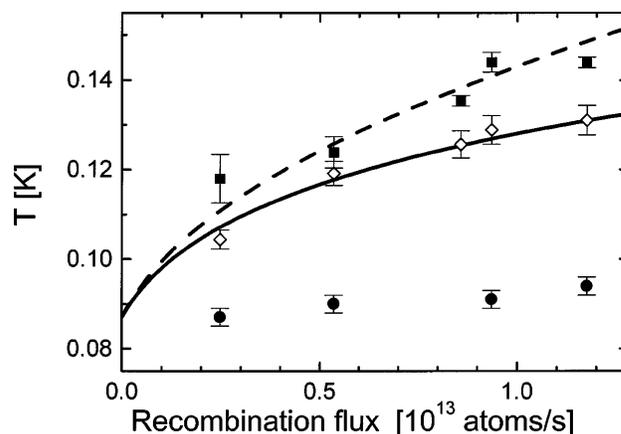


FIG. 4. Temperatures of the 3D H (squares), the He surface (diamonds), and the He bulk (circles) as a function of the recombination flux. The solid curve is the surface temperature predicted by ripplon-phonon coupling theory [13] and the dashed curve is the predicted 3D gas temperature. The He temperatures may be offset by ± 5 mK due to an uncertainty in the calibration of the thermometer.

[7]. Using the theoretical boundary resistance between ripplons and phonons [13], we calculate the expected temperature of the helium surface. The result, with no adjustable parameters, is plotted in Fig. 4.

In addition to the surface being warmer than the bulk, the 3D H gas is warmer than the surface. This is due to direct heating of the H by collisions with excited H₂. The observed heating implies an *effective* cross section of 0.007 Å² for full (4.5 eV) energy release. Arvela *et al.* [14] reported a similar value.

The heating of the surface limits the presently attainable phase-space densities to the edge of the quantum degenerate regime. Our highest degeneracy parameter is $n_2\Lambda^2 = 0.9(1)$. To extend our experiments into the degenerate regime will require a cell with more opportunity for the disposal of excited H₂ away from the recombination region. It is necessary to make the meniscus smaller to reduce the recombination area, and to change the geometry of the cell to reduce the probability for molecules to return to the meniscus. Based on our measurements of the thermal impedance, we expect that we can make these improvements while maintaining the ability to study the 2D gas optically.

Finally, we comment on the nature of the adsorbed atom spectral line. As is seen in Fig. 3 the line shift and linewidth change when we add ³He atoms. Apparently, the linewidth and line shift are proportional to the binding energy of the ground state H atoms, which is reduced from 1.01 K on ⁴He to ≈0.4 K on a surface saturated with ³He [8]. The line shift is ≈15 times larger than this binding energy. Qualitatively, this factor can be attributed to the larger polarizability of the excited atom, leading to stronger van der Waals attraction.

In Ref. [6], the possibility of the existence of long-lived bound states of the 2P atom on the He surface was examined theoretically. It was predicted that such a state would give rise to a sharp (only radiatively broadened) line at a red detuning of 180 GHz from the bulk line. The line shift is qualitatively correct, but the observed line is broader by a factor of more than 1000. This broadening cannot be dominated by nonradiative decay (predissociation), as we find the fluorescent yield (FY) to be high, FY = 0.5(1).

Also, at the densities encountered, line broadening by H-H interactions is neither expected nor observed. A possible line-broadening mechanism, suggested for alkali atoms adsorbed on helium clusters, is the reconfiguration of the helium surface after optical excitation [15]. However, in this case, nonradiative decay seems inevitable. In view of the high FY we propose another mechanism, viz., that a He-H* dimer is formed and is ejected from the surface (it can be formed in a state of ejection or it can be initially bound and eject itself from the surface while de-

exciting vibrationally). As the polarization of the light is in the plane of the surface, the dimer would be formed in the B²Π state preferentially. This state decays radiatively in vacuum [16].

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