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Thermal compression of atomic hydrogen on helium surface

24.10.2006

Keywords Atomic hydrogen · low dimensional gas · Berezinski-Kozterlitz-Thouless transition · adsorption isotherm · ripplon-phonon thermal contact

Abstract We describe experiments with spin-polarized atomic hydrogen gas adsorbed on liquid 4He surface. The surface gas density is increased locally by thermal compression up to \(5.5 \times 10^{12} \text{ cm}^{-2}\) at 110 mK. This corresponds to the onset of quantum degeneracy with the thermal de-Broglie wavelength being 1.5 times larger than the mean interatomic spacing. The atoms were detected directly with a 129 GHz electron-spin resonance spectrometer probing both the surface and the bulk gas. This, and the simultaneous measurement of the recombination power, allowed us to make accurate studies of the adsorption isotherm and the heat removal from the adsorbed hydrogen gas. From the data, we estimate the thermal contact between 2D hydrogen gas and phonons of the helium film. We analyze the limitations of the thermal compression method and the possibility to reach the superfluid transition in 2D hydrogen gas.

1 Introduction

Spin-polarized atomic hydrogen (H↓) adsorbed on the surface of superfluid helium represents an almost ideal realization of a two-dimensional (2D) Bose gas. The superfluid helium surface is a perfect substrate for the 2D gas, since it is translationally invariant and the interaction potential supports only one bound state for hydrogen atom. The main interest in studies of two-dimensional Boson gas is the formation of Bose-Einstein condensate, which is not fully understood in 2D. In contrast to a 3D condensate, the phase of a 2D condensate fluctuates on a macroscopic scale and there is no long range phase correlation in the system, while the density fluctuations are fully suppressed. The formation of superfluidity
in 2D is explained by the topological theory of Berezinskii, Kosterlitz and Thouless (BKT), and recently the BKT-type crossover has been observed in optically trapped atomic gas. Beside the Bose-Einstein condensation, reduced dimensionality can be studied e.g. in the behavior of the recombination rate constants and in the strength of interatomic interactions.

Reaching quantum degeneracy in $H_\downarrow$ is limited by the large energy release in atomic recombinations. To remove this obstacle, Kagan et al. proposed to use an open system, where most of the recombination energy is carried off by the excited $H_2$ molecules and the atoms escaping from the sample. In the case of adsorbed $H_\downarrow$ gas, only a small fraction of the energy $\sim 1\%$ remains on the surface to be removed via the excitations of the helium film, ripplons and phonons. This has been utilized in two local compression methods: thermal and magnetic compression. In the magnetic compression method used e.g. by Sazonov et al. the atoms are compressed in a small region of the sample cell (SC) wall by strong magnetic field gradients. The sample on such a “magnetic spot” is at higher temperature than the rest of the cell wall, and the recombinations heat is transferred by the ripplons along the helium surface. The cooling efficiency depends on the length of the heat path and therefore the size of the sample was reduced to about 20 $\mu$m. Due to the small size and the strong magnetic field gradient it is impossible to implement sensitive spectroscopic diagnostics of the surface atoms. In the thermal compression, first utilized by Matsubara et al., a small “cold spot” (CS) is cooled to a lower temperature than the rest of the sample cell. The compression is caused by a reduced desorption rate of the atoms from the CS, which decreases exponentially with the temperature. The heat of the recombinations is removed via the contact between ripplons and phonons of the helium film. A more detailed description of the compression methods can be found from a review of Vasiliev and Jaakkola.

In this paper we describe our experiments utilizing thermal compression to increase the density of 2D atomic hydrogen gas. General features of 2D atomic hydrogen and the compression method are found in section 2. An adsorption isotherm of $H_\downarrow$ on helium surface and a ripplon-phonon thermal contact are introduced. Section 3 contains a description of the experimental setup where we emphasize the differences between the two configurations of the sample cell used in the experiments. In section 4, the measurement procedure is explained. We describe in detail how the data are acquired and processed. In sections 5 and 6, we present the results of experiments on 2D $H_\downarrow$ and discuss the efficiency and limitations of thermal compression to reach the BKT transition.

2 Method of thermal compression

Atomic hydrogen gas is metastable against recombination into molecules. Formation of a hydrogen molecule in a collision always needs at least three participating bodies. At low bulk densities ($\lesssim 10^{15}$ $\text{cm}^{-3}$) considered the three particle collisions in the bulk are fairly rare and, therefore, the recombinations take place only in the adsorbed 2D gas phase, where the helium surface is the required third body. On the surface the recombinations proceed through the collisions of two or three hydrogen atoms. The atoms populate the lower two hyperfine states in high magnetic field and at low temperatures. These high field seeking hyperfine states are
usually denoted by letters $a$ (F=0, m$_f$=0) and $b$ (F=1, m$_f$=-1). The hyperfine energy levels are plotted in fig. 1. The state $a$ has a small admixture of the spin state with opposite orientation of the electron spin. This provides a finite recombination probability in the collisions involving $a$-state atoms and leads to a reduction of the $a$-state population to a negligible level. Then the gas becomes doubly spin polarized ($H_{↓↓}$, where $\downarrow$ marks the nuclear spin) containing atoms only in the $b$-state, which is a pure spin state. The atoms in this state can not form molecules in binary collisions, thus the decay of a doubly polarized sample is controlled by the one-body relaxation from $b$ to $a$-state and by the dipolar three-body recombination. The heat generated by the three-body surface recombination increases rapidly as a third power of density, and it is one of the major obstacles for reaching the BKT transition in $H_{↓↓}$.

An adsorption equation, e.g. a relation between the surface and bulk gas density in thermal equilibrium is found by equating the chemical potentials of the gas phases. Taking into account a possible temperature difference between the surface and bulk gas the surface density is given by

$$\sigma = \left( \frac{T_c}{T_s} \right)^{3/2} n \Lambda \exp \left( \frac{E_a}{k_B T_s} \right),$$

where $E_a$ is the adsorption energy (for H on $^4$He surface $E_a = 1.14(1) \times k_B$ K),

$n$ is the bulk gas density, $T_c$ ($T_s$) is the bulk (surface) gas temperature, and $\Lambda$ is the thermal de Broglie wavelength of the adsorbed gas. This equation is valid in non-degenerate case $\sigma \Lambda^2 \ll 1$. At higher degeneracy one has to use the full quantum isotherm taking into account Bose-statistics and interactions between the atoms.

The prefactor $(T_c/T_s)^{3/2}$ takes into account the temperature difference between the bulk and surface gases. Exponential growth of $\sigma$, proportional to $E_a/k_B T_s$, is essential for the thermal compression method. Surface density is compressed by a
factor of about 100 when the temperature is lowered, e.g., from 100 mK to 70 mK. Eq. (1) also yields \( T_s \) provided \( E_a, \sigma \) and \( n \) are known. The adsorption energy is known rather accurately from the experiments\(^{13,14} \) and it is not affected by the solid substrate beneath the liquid \(^4\)He layer unless the latter is thinner than about 20 monolayers.\(^{15} \)

About 99% of the energy generated in the surface recombination appear in the form of highly excited \( \text{H}_2^* \) molecules, which desorb from the surface and leave the recombination site. The energy is then gradually released in numerous collisions with the sample cell walls.\(^{10,16,17} \) Idea of local compression is to have a large sample cell area with small surface density through which the recombination heat is removed and only a small high density region where the recombination occurs. This can be done by creating a small cold spot having a lower temperature than the rest of the cell walls. The atoms lost in recombination on the CS, are replenished from the bulk H gas, which provides a large reservoir of H\(_{\downarrow}\) atoms and increases the lifetime of the gas.

Adsorbed hydrogen atoms are in thermal equilibrium with the ripplons of the helium surface. This is ensured by the fast momentum relaxation of adsorbed atoms caused by the interactions with ripplons. At 100 mK the characteristic relaxation time \( \tau_p = 3 \times 10^{-8} \) s is much shorter than the surface residence time \( \tau_r \) of hydrogen atom on liquid \(^4\)He.\(^{18} \) The recombination heat accumulated into the system of H+ripplons is transferred to the sample cell body through the phonons of the helium film and has to pass through the ripplon-phonon thermal contact. The thermal coupling of H to phonons of the film is rather weak.\(^{10,16,17} \) The maximum cooling power calculated for the phonon system at temperature \( T_{ph} \) is given by\(^{19} \)

\[
P_{rp} = G_{rp}(T_s^{20/3} - T_{ph}^{20/3}),
\]

where \( G_{rp} = 0.84 \) Wcm\(^{-2}\)K\(^{-20/3}\) is the heat conductance of the ripplon-phonon contact. The cooling power is strongly temperature dependent and \( G_{rp} \) is smaller than the Kapitza conductance across, e.g., metal/\(^4\)He interfaces below 100 mK.\(^{20} \)

3 Experimental

Experiments were carried out in a top-loading cryostat with two home-made refrigerators, a dilution refrigerator (DR) and a \(^3\)He refrigerator. The latter has been constructed to cool a cryogenic rf dissociator, operating at 600-800 mK. The dissociator contains a helical 350 MHz resonator and operates in a pulsed mode. The construction of the dissociator is described elsewhere.\(^{21} \) We were able to obtain a hydrogen flux reaching the sample cell SC in the range \( 10^{11} - 3 \times 10^{13} \) atoms/s by varying the pulse width (0.2-1 ms), the repetition rate (10-100 Hz) and the rf power fed into the dissociator (1-10 mW). The rf discharge also leads to evaporation of the helium film in the dissociator, and the helium vapor flows down and recondenses in the filling line. To prevent the vapor condensation in the sample cell we installed an accommodator into the filling line just below the magnet fringe. The accommodator is thermally anchored to the 0.3 K heat exchanger cooled by the dilute stream entering into the still of the DR. After the accommodator the H\(_{\downarrow}\) atoms pass through a hyperfine state polarizer, which is designed to remove the
Fig. 2 Cooling of the cold spot and the sample cell. The cold spot temperature is set by the cooling liquid temperature. A stable operation of the refrigerator is maintained by the by-pass loop in the diluted stream.

\( a \)-state atoms. The polarizer is a cylindrical chamber with several baffles inside to increase its surface area. It is located in \( \approx 3.2 \) T field and thermally anchored to the lowest heat exchanger of the DR operating at a temperature range from 100 to 120 mK. The temperature and magnetic field in the polarizer are optimized to burn most of the \( a \)-state atoms before they enter into the sample cell, and this way we obtain the maximum surface density on the CS in the steady flux experiments as described below.

The sample cell is located in the center of a superconductive magnet operating at 4.6 T and is thermally anchored to the mixing chamber (MC) of the DR, which has the base temperature of 30 mK and the cooling power of 150 \( \mu \)W at 100 mK. Due to a relatively long distance of about 40 cm from the MC to the SC a mechanical thermal anchor is not convenient for cooling a small cold spot. Therefore, we utilized the dilute \(^3\text{He} - ^4\text{He}\) “coolant” mixture from the mixing chamber of the DR to cool the CS. Helium mixture is a perfect dielectric and can fill up the mm-wave resonator without disturbing its Q-value. At a typical circulation rate of 100 \( \mu \)moles/s the mixture flows with the speed of 2 cm/s in a 1.5 mm diameter tube and it has relatively large specific heat. Another advantage of this method is a possibility of rapid (2-3 sec) change of the coolant temperature \( (T_l) \) with a heater attached to the coolant tube going to the cell. If the whole dilute stream of the DR would be used to cool the cold spot, then, at high \( T_l \), the operation of the DR would be seriously disturbed. Therefore, the stream is divided into two approximately equal parts as illustrated in fig. 2. Half of the stream flows through a by-pass loop and merges the CS stream before entering into the DR heat exchanger. With this construction we can raise \( T_l \) up to 300 mK while still being able to keep the SC and MC temperatures below 50 mK. Temperatures of
Fig. 3 Left: The SCI with 6 mm diameter cold spot located on the lower surface of the Mylar film forming the upper wall of the SC. Right: The Fabry-Perot resonator and the cold spot in the SCI. The contour plot shows the mm-wave field intensity profile of TEM$_{003}$ mode.

Electron-spin resonance (ESR) at 129 GHz is used to detect the hydrogen atoms. The bulk and surface adsorbed atoms are in contact with resonant mm-wave field in an open Fabry-Perot resonator (FPR) similar to the optical resonators used in lasers. Open geometry was chosen to ensure an easy escape of the recombination products from the CS, which is crucial in 2D compression experiments. Schematic drawings of the sample cell with the FPR and CS is shown in fig. 3. The upper mirror of the resonator is hemispherical with 6.5 mm radius of curvature, while the lower mirror is flat. The spacing between the mirrors is ≈ 5.1 mm corresponding to the TEM$_{003}$ operating mode with loaded $Q \approx 1700$. The resonator space is split in two parts, isolating the SC from the vacuum space, by a 20 µm Mylar film glued on the top of the sample cell body. A Stycast 1266 ring, glued between the semispherical mirror and the Mylar foil as shown in fig. 3, seals the coolant volume. The coolant flows in and out of the resonator through 1 mm i.d. copper tubes soldered into the semispherical mirror. The coolant stream flushes the upper surface of the Mylar foil and thus creates a 6 mm diameter cold spot on its lower surface. The space in the resonator beneath the foil is filled by the hydrogen bulk gas. This construction gives a good thermal isolation between the CS and the SC, and makes it possible to use SC temperatures of about 200 mK without a significant heat leak to the CS. Also, the thermal boundary resistance between helium and Mylar is one of the lowest of all solids, which provides the best cooling of the CS. During operation the FPR frequency is slightly dependent on temperature. This is because, three quarters of the resonator is filled with $^3$He/$^4$He mixture whose dielectric constant is a function of the concentration ratio, which is temperature dependent. Fortunately the dependence is rather weak...
at $T_i \lesssim 100$ mK and does not disturb noticeably the stability of the ESR signal. On the other hand, by heating the coolant to temperatures of the order of 1 K leads to an ESR resonator frequency shift of about 50 MHz and helps to find the resonance frequency after the cool down from room temperature.

The size of the gap between the cold spot and the lower FPR mirror is very important because it influences the escape of the recombination energy from the CS. In the first version of the sample cell described above, the gap size was $\approx 1.1$ mm, and from the measurements (see Sec.4 and 5) we concluded that there is a significant recombination overheating of the central part of the CS. To reduce the overheating we have modified the FPR construction. The lower mirror was replaced with hemispherical one, and moved down by $\approx 2.5$ mm increasing the gap to the CS by a factor of $\sim 3$ and changing the resonator mode to TEM$_{005}$. As we demonstrate below, this led to a substantial improvement of the heat removal from the CS. In this paper we present experimental results obtained in both versions of the sample cell, further on referred as SCI and SCII.

Another important parameter in the thermal compression experiments is the size of the cold spot. In our previous work $^{11,24}$ we used 1.5 mm diameter cold spot. This size has been chosen to minimize the total three-body recombination rate and avoid overheating of the sample cell. It was about twice smaller than the characteristic width of the mm-wave profile (beam waist dia.$\approx 3.2$ mm), which allowed imaging of the density profile of the adsorbed gas near the edge of the CS.$^{11}$ In the experiments we found that the three-body recombination rate constant is $\approx 20$ times smaller than it was anticipated on the basis of existing data. The contribution of the cold spot to the total loss rate was negligibly small and we were not able to study recombination processes on it. Therefore in the present setup we increased the CS diameter to 6 mm. With this size most of the recombination occur on the CS, and we were able to perform an accurate measurement of the three-body recombination rate constant.$^{25}$ With the large spot we lost the possibility to image the density distribution near the CS edge, which went beyond the sensitivity region of the FPR. On the other hand, probing the adsorbed gas at uniform density offers a possibility to study the effects of homogeneous ESR line broadening caused by interatomic collisions, provided that other broadening mechanisms are reduced to a negligible level.

Accurate measurements of the recombination heat released in the sample requires that all the energy is dissipated in the SC, and nothing is transferred back to the filling line by the excited H$_2$ molecules. To prevent such a heat leak we installed a $^4$He fountain valve into the H filling line at the SC bottom as shown in fig. 2. The valve is controlled by a heater and the helium level is measured with a capacitive level gauge.

The rate of the impurity-induced $b - a$ relaxation depends on the quality of the sample cell walls. To inhibit this unwanted process we followed the standard procedure in the hydrogen research. The SC was made of high-purity Cu and carefully etched in order to reduce the relaxation. After that, we covered the inner surfaces of the cell with polymer foil and epoxy. The impurity relaxation was further decreased by building up a layer of solid H$_2$ on the SC walls. The layer is grown slowly during a week when the H$_2$ flux to the SC is kept on continuously.$^{26}$ All these measures decreased the intrinsic one-body relaxation rate to a negligible level when compared to the three-body recombination rate on the CS.
The ESR spectrometer is based on a home-made 129 GHz heterodyne receiver and its cryogenic part is thermally anchored to the 1 K pot of the DR. The spectrometer is capable of detecting both absorption and dispersion signals simultaneously and has a sensitivity of about $10^9$ spins/G at mm-wave excitation power of 20 pW.

4 Measurement procedures and density calibrations

Two types of measurements have been used to study $\text{H}_2$. In the decay type measurements the hydrogen sample is first accumulated into the sample cell, the dissociator is turned off and the experimental conditions (temperature, volume or pressure) are fixed. Then, the natural decay of the gas sample is monitored. With this method most of the recombination mechanisms has been studied. In the second method, hydrogen source is running and filling the sample cell continuously, so that a steady state is reached. The equilibrium of the gas is balanced by incoming flux and recombination. Main advantage of this method is a long term stability of bulk and surface density and an easy control of recombination power by the incident flux. In the present work most of the measurements were of the decay type. The steady flux method was used only for measuring the ripplon-phonon thermal contact as described at the end of this section.

In the beginning of an experimental run a small amount (typically 1-4 mmoles) of $\text{H}_2$ is loaded into the dissociator from room temperature. The $\text{H}_2$ gas freezes on the dissociator walls and resonator helix. After that, we condense $\approx$ 8 mmoles of isotopically purified $^4\text{He}$ ($\leq$ 1 ppb of $^3\text{He}$) into the sample cell, required for proper operation of the fountain valve. Superfluid helium film is formed lining the inner walls of the dissociator and the SC. For the decay measurements we run the dissociator at average power of 2-3 mW which is limited by the back flow of helium into the dissociator along the filling line walls. The highest flux obtained was $2 \times 10^{13}$ atoms/s entering the sample cell. It usually takes about 2000 s to reach the saturation bulk density of about $2 \times 10^{15}$ cm$^{-3}$. The hyperfine polarizer does not work at high dissociator power, and the SC and MC are overheated to $\approx$ 250 mK due to the $a-b$ recombination and re-condensing helium vapors in the filling line and the SC. After switching off the dissociator we cool down the mixing chamber, sample cell and CS coolant to desired temperatures below 180 mK, which usually takes 5 to 10 min. During this time the $a$-state atoms are burned in recombination and the $\text{H}_2^+$ sample is formed. Typically, we start the decay measurements with the bulk density of $1-5 \times 10^{14}$ cm$^{-3}$ and we are able to reach the maximum surface density of $5.5 \times 10^{12}$ cm$^{-2}$ on the CS. The feedback power of the SC temperature controller and the ESR spectra of bulk and surface atoms are the data collected during the decay measurement. The raw data are presented in figs. 4 and 5.

The ESR line of the adsorbed atoms is shifted from that of the bulk gas due to the internal dipolar field created by the polarized atoms in the plane where they are adsorbed. When the plane is perpendicular to the polarizing magnetic field the dipolar field is proportional to the surface density as $\Delta H_d = -1.1(1) \times 10^{-12} \sigma$ G cm$^{-2}$. Due to rapid motion of the atoms, fluctuations of the dipolar field are well averaged and the intrinsic linewidth of the surface gas is expected to be very small. In practise the linewidth is determined by the inhomogeneities of
the polarizing magnetic field and/or surface density inhomogeneities. The latter is especially important in the thermal compression experiments due to an inhomogeneous temperature profile near the cold spot edge.\textsuperscript{31}

The ESR line integrals of both the surface and bulk atoms are proportional to the density of atoms. The absolute density calibration is not an easy task, since we cannot employ calibration samples which are typically used for this purpose in ESR spectroscopy. The calibration of the H\textsubscript{\textsuperscript{\textleftarrow{\textdownarrow}}} bulk density is made calorimetrically by measuring the recombination heat released in the sample. We found this method more simple and reliable than the calibration based on the absolute measurement of microwave characteristics of the ESR resonator: coupling parameter and Q-value.\textsuperscript{30} The SC temperature controller adjusts the heating power \( P \) so that the sample cell temperature is constant. A decrease or increase in the recombination power is compensated by the temperature controller. In fig. 5 the recombination power \( P \) is shown for several decays at different temperatures of the CS cooling mixture. In these experiments the number of atoms in the bulk of the sample cell is much larger than on the surface. Then, by integrating the recombination power, we calculate the bulk density

\[
n(t) = \frac{2}{V_h D} \int_0^\infty [P_0 - P(t')] dt',
\]

where \( V_h = 40(1) \text{ cm}^3 \) is the volume of the sample cell, \( D = 4.478 \text{ eV} \) is the energy released in a single recombination event,\textsuperscript{31} and \( P_0 \equiv P(\infty) \) is the stabilization power at \( n = 0 \).

Similar procedure cannot be used for measurement of the surface density because the number of surface atoms in the ESR resonator is negligibly small...
Fig. 5 The heating power $P(t)$ applied to sample cell by the temperature controller at $T_c = 178$ mK and $T_l$ ranging from 98 mK to 162 mK. The linear fit to the flat part of the signal gives a background heating power $P_0 = 20.3 \mu$W for $T_l = 129$ mK. For the lowermost trace the time scale is multiplied by 2.

compared with that of the bulk gas. It is known that the strength of the bulk ESR signal is proportional to the product $n \cdot \int V H_1^2 dV$ with $H_1$ being the magnetic component of mm-wave excitation and the integral is taken over the volume occupied by hydrogen gas. The surface line strength is, respectively, proportional to $\sigma \cdot \int A H_1^2 dA$ where the integral is taken over the area of the CS. To evaluate the ratio of the integrals we solved numerically the mm-wave field profile in the Fabry-Perot resonator. Once the ratio is known the surface density can be extracted by using the calorimetrical calibration of the bulk density. Main errors of the surface density measurement arise from the inaccuracy of the mm-wave field calculation (\sim 5\%) and from the drifts of the ESR spectrometer excitation power and sensitivity caused by the changes of liquid helium level in the cryostat. We estimate that the overall error in $\sigma$ extracted from the ESR integrals is about 10\%.

Decays of $H_{1\parallel}$ in the experimental conditions considered in this work are controlled by two processes: one-body a-b relaxation and three-body recombination on the walls of the SC and CS. They can be easily distinguished by changing the CS temperature as demonstrated in fig. 6. At high enough temperature of the CS coolant ($T_l = 133$ mK data in fig. 6) surface density on the CS is low and the influence of the three-body recombination is negligibly small; thus the density decays exponentially as a function of time. The slope of the decay curve in semi-log plot gives $G_1^\parallel$, the one-body relaxation rate. Cooling of the CS strongly increases the loss rate ($T_l = 78$ mK data in fig. 6), which is mainly caused by the three-body
Fig. 6 Bulk densities extracted from the ESR spectra at $T_c = 178$ mK. □ - $T_l = 133$ mK and ◦ - $T_l = 78$ mK with the SCI and △ - $T_l = 78$ mK with the SCII. The solid line with error bars is from a calorimetric density calibration and the dashed line is an exponential fit to ESR data points. The decays at $T_l = 78$ mK are not exponentials because of the high three-body recombination rate on the CS.

In a general case the recombination rate can be found as

$$ \frac{dn}{dt} = V_h G_e n + A_s L_3^s \sigma^3, $$  \hspace{1cm} (4)

where $L_3^s$ is the surface three-body recombination rate constant and $A_s$ is the area of the cold spot. By subtracting the one-body contribution, extracted from the high $T_l$ data, it is possible to obtain the value of the three-body recombination rate constant $L_3^s = 2.0(7) \times 10^{-25}$ cm$^4$ s$^{-1}$.

In steady flux measurements the sample cell is continuously filled with atoms. At the maximum dissociator power the hyperfine polarizer is strongly overheated and the SC is being filled with atoms in both $a$ and $b$ states in almost equal amounts. The total recombination power is so high that we can not get the surface density on the CS higher than $10^{12}$ cm$^{-2}$. When the dissociator power is decreased by about 20% we get the polarizer cold enough, and it effectively burns off most of the $a$-state atoms before they enter the SC. Then, a maximum steady state surface density of $3.5 \times 10^{12}$ is reached with a bulk density of $n \sim 10^{13}$ cm$^{-3}$. Further decrease of the dissociator power leads to a decrease of the steady state surface density. With the constant flux method we measure steady state bulk and surface densities as functions of total recombination power. Although we can not reach high quantum degeneracy with this method, it allows to study the transfer of recombination heat through the helium surface.
Fig. 7 Surface density plotted as a function of bulk density for several decays at different \( T_c \) and \( T_l \). The surface temperatures \( T_s \) are extracted by fitting the adsorption isotherm to the linear parts of the data (solid lines).

5 Results and discussion

For an estimate of the quantum degeneracy reached in the experiments we need to known the true temperature of the surface gas \( T_s \). There is no direct means to measure it, but it is possible to evaluate \( T_s \) from the adsorption isotherm eq. (1) using the known values of the bulk and surface gas densities. In fig. 7 we present \( \sigma \) as a function of \( n \) measured at different coolant temperatures \( T_l \). At high enough CS temperature we are in the limit when the recombination overheating is negligibly small, and \( T_s \) remains constant during the decay. Then, the surface density increases linearly with \( n \) according to eq. (1). This regime is seen in fig. 7 from the data with \( T_l = 129 \) mK. The slope of \( \sigma(n) \) is proportional to \( \lambda(T_c/T_s)^{3/2}\exp(E_a/T_s) \), and using the value of \( E_a/k_B = 1.14(1) \) K we calculated the surface gas temperature \( T_s = 130 \) mK. This is, within the accuracy of our thermometry, the same as the coolant temperature, which confirms the reliability of the \( T_s \) determination and the absence of overheating. This measurement is the first experimental check of the adsorption isotherm in the case of different bulk and surface gas temperatures. At lower coolant temperatures \( \sigma(n) \) is not linear any more and gets saturated at higher \( n \). One can see in fig. 7 that the saturation starts at lower \( n \) with the lower coolant temperatures.
First we consider only the low density region of the data, where \( \sigma(n) \) is linear. The linearity of \( \sigma(n) \) even for the lowest \( T_l \) implies that the surface temperature \( T_s \) from the adsorption isotherm is constant. However, the difference between \( T_s \) and \( T_l \) is larger, the lower the temperature \( T_l \) is. We consider two possible explanations for this effect. (i) The CS surface is overheated by the heat leak from the warmer CS walls along the helium film. (ii) The 2D gas is not in equilibrium with the bulk gas due to a hydrodynamic flow along the surface. The flow is driven by the gradient of 2D pressure at the edge of the CS. If the adsorption-desorption exchange between the bulk and surface gas \( \sim n \) becomes slow enough, the surface density on the CS may be substantially smaller than the equilibrium value. Then \( T_s \) extracted from the adsorption isotherm is higher than the true surface temperature.

In the limit of ultra low density, when there is practically no flux of atoms from the bulk, the 2D pressure \( P \sim n \cdot T \) is constant over the surface and thermal compression cease to work. Measurements in this regime would give a nice possibility to explore the equation of state of a 2D gas, which would be especially interesting at quantum degeneracy. However, we found that an increase of the temperature difference between the SC and the CS coolant (e.g. from \( T_c - T_l = 73 \) mK to 100 mK in fig. 7) leads to an increase of the effective surface temperature \( T_s \). This confirms qualitatively that the CS overheating at low \( \sigma \) is caused by the heat flux from the SC. Also the flow of adsorbed atoms out of the center of the cold spot would create a nonuniform density profile with the density decreasing from the center of the CS towards its edge. This would be observed as a broadening of the left wing of the ESR lineshape, which was seen previously on the smaller size cold spot. In the present setup we have not observed such a broadening. Also, the analysis of the hydrodynamic flow is complicated due to the gas of ripplons on the helium surface. The effects of the flow are expected only at very small bulk and surface densities, which are beyond the sensitivity of our apparatus and thus making their experimental study impossible at present. Therefore, in the following we concentrate on the high density data, where the main goal of this work, high quantum degeneracy, can be reached.

At high surface density the 2D gas is overheated, which is revealed in the leveling off of \( \sigma \) with increasing \( n \) in fig. 7. This is mainly caused by the three-body recombination. At the same time in the SCI we observed an abrupt increase of the surface ESR linewidth (FWHH), as plotted in fig. 8. The surface ESR line changed from close to Lorentzian shape to asymmetrical shape with strongly broadened right wing. To understand the nature of the line broadening, we studied how does the lineshape depend on the magnetic field gradients \( \frac{\partial B_z}{\partial r} \) created by a special shim coil around the SC. By changing the direction and the magnitude of the current in the coil we can compensate most of the inhomogeneities in the internal dipolar field of the 2D gas. We found that the broadening can be substantially reduced by the extra magnetic field with a parabolic radial profile and having the minimum in the center of the CS. This means that the internal dipolar field and the surface density profile have the minimum in the center of the cold spot. This can be explained by the presence of an extra heat source in the center of the CS, decreasing towards the edge.

As already mentioned in sec. 2, it is very important in the compression experiments to ensure free escape of the recombination products from the compression region. Although the geometry of the Fabry-Perot resonator was designed to be
open, the $H_2^*$ molecules formed on the CS are not completely free to leave the CS. The flight path is restricted by the lower ESR mirror below the CS. The molecules bounce from the lower mirror and may return back to the CS. It is known that $H_2^*$ molecule needs approximately 150 surface collisions with the helium film to release half of its internal energy. The average $H_2^*$ excitation energy released in a single wall collision is then about $170 \, \text{K} \times k_B$. The presence of the lower mirror increases the average number of collisions in the center of the CS compared that of near the edge and may lead to the observed inhomogeneous overheating of the CS surface.

We performed a numerical simulation to estimate the extra heat flux due to the reflecting molecules. In the simulation we assumed that every molecule has a random recombination site on the CS and a random direction of flight. The collisions with the CS and with the flat mirror were considered to be elastic as illustrated in fig. 9. After reaching the edge of the mirror the molecule escapes and a new molecule is generated. We calculated $N_{\text{col}}$, the average number of collisions per unit area with the CS surface per number of molecules generated, as a function of the distance to the spot center (fig. 9). For the geometry of the SCI with the flat ESR mirror being 1 mm below the CS, the simulation gives parabolic $H_2^*$ flux profile with $N_{\text{col}} = 2.4 \, \text{cm}^{-2}$ in the center and decreasing by a factor of 1.7 at the edge of the CS. This creates a heating of about $120 \, \text{K} \times k_B \, \sigma \, \text{cm}^{-2}$, about half
Fig. 9 Scheme of the CS overheating in SCII and the calculated flux of H$_2^*$ collisions per recombination event reflecting from the flat mirror. The upper and the lower curve corresponds to different distances $d$ between the flat mirror and the CS.

of the recombination heat delivered directly to the surface when the molecules are created. If the gap between the mirrors is increased to 2.7 mm, the collision profile becomes almost flat and the number of collisions is about 0.5 cm$^{-2}$. This is still substantially larger than that for a completely open cell where $N_{col} = A_c^{-1} \approx 0.01$ cm$^{-2}$ and $A_c$ is the area of the SC. Once the reason for the overheating was realized we modified the sample cell. The lower mirror was moved down increasing the gap under the CS to 2.7 mm in the SCII. We also had to replace the flat mirror with a hemispherical one in order to have a working resonance mode at so large spacing (TEM$_{006}$ mode). With these changes we eliminated the line broadening effect, as demonstrated in fig. 8 and succeeded to increase the maximum surface density from $3.6 \times 10^{12}$ cm$^{-2}$ to $5.5 \times 10^{12}$ cm$^{-2}$.

If the flat mirror in simulation is replaced with the spherical mirror of SCII, the number of collisions on the CS increases by an order of magnitude due to focusing effect of the spherical mirror. This is in contradiction with the experiments with the SCII, where the heating of the CS is clearly reduced. This means that the collisions of the molecules with the helium film cannot be assumed to be completely elastic. The inelastic collisions are not taken into account in the simulation, which may be the reason why the model fails to work for semispherical mirror.
The thermal contact between the ripplons and phonons of helium film $G_{rp}$ turns out to be a bottleneck of the recombination energy removal from the cold spot. To measure this quantity one needs to know the heat flux and both the phonon and ripplon temperatures. In this experiment we used the constant flux method. By measuring the bulk and surface densities we extracted the ripplon temperature $T_s$ as described above. There is no means to measure the temperature of the phonons of the helium film, but since the thermal contact between the helium film and the coolant is much better than the ripplon-phonon contact, the phonon temperature is close to $T_l$. Furthermore, if $T_l \ll T_s$, the $T_l^{20/3}$ term in eq. (2) is negligibly small making the accurate determination of $T_l$ unnecessary. In the constant flux method, all the atoms entering the sample cell will recombine there in the exchange $a$-$b$ recombination, thus, the total loss rate is equal to the input flux and can be adjusted by the dissociator power. However, to know the heat flux through the CS surface, we have to make sure that the recombination energy is evenly distributed over the cell walls and the CS. This is not the case if the CS temperature is much lower than the cell temperature. Therefore, in the $G_{rp}$ measurements, we kept the CS and SC temperatures rather close to each other, so that the rate of the $a$-$b$ recombination on the cell walls is much larger than that on the cold spot, i.e. $A_c \exp(2E_a/T_c)/A_s \exp(2E_a/T_s) \gg 1$ with $A_c$ and $A_s$ being, respectively, the areas of the SC and the CS. Then the share of the total recombination power absorbed by the CS is equal to $P_{rp} = P \cdot A_s/A_c$, with $P$ being the total recombination power measured by the cell temperature controller. In fig. 10 we plot the heat flux through the unit area versus the surface temperature $T_s$ for two different coolant temperatures. Clearly the cooling power of the CS is strongly dependent on the difference between $T_s$ and $T_l$. The data of fig. 10 were fitted to eq. (2) with $G_{rp}$ and $T_l$ as free parameters. The fit gives $G_{rp} = 7 \pm 2$ W/cm$^2$ and $T_l$ values deviating less than 1 mK from the thermometer reading. The result for $G_{rp}$ is about 8 times larger than the theoretical value. The discrepancy may arise from our assumption that all the recombination heat is dumped to ripplons. A large fraction can be transferred directly to phonons or rotons of the helium film via the recondensing helium atoms. Therefore our result can be considered as an upper limit estimate of $G_{rp}$. To our knowledge, this is the first measurement of the ripplon-phonon thermal conductivity.

6 Limitations and future prospects

In this work the highest surface density observed was about $5.5 \times 10^{12}$ cm$^{-2}$ at $T_s \approx 110$ mK, which corresponds to the 2D phase-space density $\sigma \Lambda^2 \approx 1.5$. The dense surface gas was overheated well above the coolant temperature and $\sigma$ was almost independent of the bulk gas density as shown in fig. 7. The main source of heat in the SCII was the heat transferred to the CS surface directly from surface recombination. Taking into account only the heat of direct three-body surface recombination, a simple equation for the maximum quantum degeneracy $\sigma \Lambda^2$ on the CS can be derived. The recombination heat absorbed by the surface of the cold spot is given by

$$P_{3b} = (f + (1 - f) \frac{A_s}{A_c}) \frac{D}{2} L_s \sigma^3,$$

(5)
The heating power, $P_h$, is plotted in Fig. 10 as a function of surface temperature, $T_s$. Different symbols correspond to different coolant temperatures: $+T_l = 85$ mK and $\times -T_l = 88$ mK. The lines are fits of the data to eq. (2).

where $f$ is the fraction of the heat dumped directly to the cold spot surface ($f \leq 2\%$). The efficiency with which this heat can be removed is defined by the ripplon-phonon thermal contact (eq. (2)). Setting $P_{3b} = P_{rp}$ we get an estimate of the highest achievable quantum degeneracy as

$$\sigma_2 \equiv \sigma \Lambda^2 = \frac{2\pi \hbar^2}{mk_B} \sqrt{\frac{2G_{rp}}{DL_3(f + (1 - f)A_{scs})T_s}} \approx \frac{1}{2} T_s^{1/2},$$

where $G_{rp} = 7 \text{ Wcm}^{-2}\text{K}^{-20/3}$ has been used to get the last form. In fig. 11 we present the results of thermal compression experiments with the highest quantum degeneracy together with the plots of the adsorption isotherm at several surface temperatures. Two sets of data are plotted, one with SCI and the other with SCII. The result of the improvement can be clearly seen as a factor of 1.5 increase in the surface density. The dashed line marks the condition of quasi-condensate formation $\sigma_2 = 3$. We include an abrupt change of the interactions in 2D gas, which leads to an increase of $\sigma$ by a factor of two in the quasi-condensate. Eq. (6) together with the adsorption isotherm defines the maximum surface density $\sigma(n)$, which can be reached for a given bulk density $n$ and temperature $T_c$. For $T_c = 178$ mK we plot $\sigma(n)$ in fig. 11 as the thin solid lines. The lower line corresponds to the theoretical value of $G_{rp}$, and the upper line is calculated using the experimental value obtained in previous chapter. The SCII data show that $\sigma(n)$ is higher than it is possible according to the theoretical $G_{rp}$ value, but the values are still well below the value calculated with the experimental $G_{rp}$. This can
**Fig. 11** Thick solid lines: Calculated isotherms of the surface gas. Dashed line: Quasi-condensate formation at $\sigma A^2 = 3$. Thin solid lines: Values calculated from eq. (6). The lower line is calculated by using the theoretical value of $G_{rp}$ and the upper line by using the experimental value obtained in this work. Experimental data ($\circ$) and ($\Box$) are measured at $T_l = 67$ mK and $T_c = 178$ mK respectively with SCI and SCII. The point ($\triangledown$) corresponds to the maximum 2D quantum degeneracy achieved with magnetic compression.

indicate that even in the SCII the CS is overheated by the reflected molecules. Thus, by removing the molecules, the density can be increased close to the quasi-condensation limit. This can be done by replacing the lower spherical mirror with a grid or mesh, allowing for most of the excited molecules to fly through the mirror. The best solution would be to make the cold spot area completely open. However, this is difficult to combine with the detection of atoms using a mm-wave resonator.

Another possibility to increase $\varpi$ according to eq. (2) is to use higher $T_s$. As one can see in fig. 11, the maximum achievable degeneracy goes well above the KTT line at higher temperatures. This is because the ripplon-phonon thermal conductance grows rapidly with increasing temperature. However, the use of higher temperatures at the same time requires a large increase of bulk density, which would, e.g., at $T = 200$ mK approach to $10^{17}$ cm$^{-3}$. At so high bulk densities one will come across with thermal accommodation heating the surface of the CS. The heating power of thermal accommodation is given by

$$P_{\text{acc}} = \frac{n v s}{4} 2k_B(T_c - T_s),$$

where $v$ is the thermal velocity and $s$ is the sticking probability of H atom to the $^4$He surface. The heating $P_{\text{acc}}$ becomes comparable with the surface three-
body recombination heat flux if the temperature difference exceeds 100 mK at \( n = 6 \times 10^{16} \text{ cm}^{-3} \). Therefore, a further increase of \( n \) does not help to reach a higher degeneracy of the surface gas. The thermal accommodation heat can also explain why the highest \( n \) data in fig. 11 deviate from the prediction of eq. (6).

The adsorption energy is one more parameter which one may try to change in order to get a higher degeneracy. Increasing \( E_a \) together with the temperature, so that the ratio \( E_a/T_s \) remains constant, will allow to improve the ripplon-phonon contact and keep the bulk density low enough. The adsorption energy can be increased by utilizing a thin undersaturated helium film at the CS. Working with the thinner films may also have several other benefits. The rate constant of the three-body recombination should decrease at higher \( E_a \). Also, one may hope on some improvement of the surface cooling due to a direct interaction of the ripplons with the phonons of the solid substrate. Making a superfluid helium film locally thinner is, however, a difficult cryogenic problem with no ready solution known at present.

Acknowledgements We would like to thank Michael Hayden, Matti Krusius, Simo Jaakkola, and Kalle-Antti Suominen for illustrative discussions. This work was supported by the Academy of Finland and the Wihuri Foundation. J. J. acknowledge support from the National Graduate School in Materials Physics. J. A. acknowledge support from the National Graduate School in Nanoscience.

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