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Chemical reaction of surface state electrons on liquid helium with atomic hydrogen

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Abstract

Chemical reaction in a two-dimensional mixture of electrons and atomic hydrogen (H) on the surface of superfluid ^{4}He was observed. By monitoring the conductivity of surface state electrons with Corbino electrode, small amount of H atoms were introduced. Hydrogen destroyed the conductivity signal and we believe it is because of electron capture reaction, $H + e^- \rightarrow H^-$. The reaction rates increased as the temperature decreased and it saturated below 200 mK. This temperature dependence agrees with our analysis which assumes that the chemical process takes place only in the surface state. © 2000 Published by Elsevier Science B.V. All rights reserved.

Keywords: Surface sate electrons; Atomic hydrogen; Superfluid helium; Electron capture reaction; Negative hydrogen

1. Introduction

A cryogenic atomic hydrogen (H) system shows quantum properties because of its small mass [1]. Since it remains gaseous down to absolute zero temperature, it behaves as a quantum gas. One of the main goals in this field was to achieve Bose–Einstein condensation and it was recently realized in a magnetic trap by the MIT group [2]. Another remarkable property is that H atoms are adsorbed on the surface of liquid helium to form a two-dimensional (2D) Bose gas. It forms a 2D quasicondensate at sufficiently high density and low temperature and observation of this state was reported [3].

Surface state electrons (SSE) also form a 2D system on liquid helium. It provides many interesting properties, such as Wigner solid transition, transport phenomena, etc. [4]. Moreover, SSE can serve as a sensitive tool for studying the superfluid helium surface, since the motion of SSE is affected by elementary excitations in underlying liquid (for example, Ref. [5]).

The question we study here is: What occurs when SSE are exposed to H? From the analogy that two H atoms recombine into molecular state at low temperatures [1], we can expect a chemical reaction between H and electron (e^-), $H + e^- \rightarrow H^- + 0.75$ eV, namely, electron capture reaction, on the surface.

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The product, negative hydrogen ion H^-, is interesting, because it is very different from other two electron atom/ion, such as He, Li^+, etc. because of the strong repulsive correlation between two electrons compared to the attractive interaction of electron with the nucleus [6]. H^- has only one bound state [7]. The reaction at low temperature has a purely quantum nature. In addition, it is of great interest whether H^- stays on the surface or penetrates into the bulk liquid. If it floats on the surface, we can obtain a new 2D charge system on liquid helium.

As far as we know, there is no prior work on the mixture of H and e^- or on H^- ion at low temperatures. Since H^- plays important roles in the atmosphere of the Sun or other stars or in accelerators, most of the works have been concentrated on the excited states of H^- and high-energy collision between H and e^- [7].

We report here the first experimental study of the 2D H–e^- mixture on the surface of superfluid ^{4}He, at temperatures from 0.1 to 0.4 K. We introduced H atoms to the surface where SSE were prepared. The chemical reaction between H and e^- does take place in the adsorbed phase, we believe it is the electron capture reaction by H.

2. Experimental

In the sample cell, a Corbino electrode is placed to measure the conductivity of the SSE. It is made from copper-coated epoxy plate by etching. The diameters are 25 mm for the outer ring and 17.6 mm for the inner disk. The gap between these two is 0.1 mm. The details and electronics for conductivity measurement are described in Ref. [8]. The distance between the upper and lower (Corbino) electrode is 3 mm and the liquid ^{4}He is prepared such that the surface is at 1 mm above the Corbino electrode. SSE are obtained by trapping the thermoelectrons emitted from a tungsten filament. It is important to ignite the filament at about 1.5 K where a certain density of helium vapor exists, in order to reduce the energy of the electrons by collisions with the atoms in the vapor before arriving at the surface. Otherwise, the energetic electrons penetrate into the liquid. A positive DC bias voltage is applied to the Corbino electrode relative to the upper electrode and the guard ring. This voltage creates electric field which holds the SSE at the liquid surface.

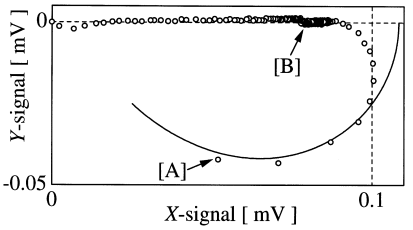

Fig. 1. Argand diagram of the conductivity signal. The open circles represent the measured signal and the solid line shows the calculated curve. The surface was charged at [A] and H atoms were introduced at [B]. The deviation from the solid curve between [A] and [B] is due to the shift of the liquid level in the sample cell during the cooling down process.

H atoms are produced by cryogenic-pulsed RF glow discharge, which dissociates H_2 molecules. The dissociator is thermally anchored to the still of the dilution refrigerator, the temperature of which is 0.7–0.8 K. A helical resonator tuned at 439 MHz is in the dissociator and its quality factor is $Q = 800$. H atoms are delivered to the sample cell through 0.7 mm i.d. Cu–Ni capillary.

The conductivity signal of Corbino electrode is shown in Fig. 1. The inphase (x) and quadrature (y) output signals of lock-in amplifier are plotted as horizontal and vertical axes, respectively, which is known as Argand diagram [8]. The solid line is a calculated curve. As for constant surface charge, the trajectory of the signal moves from the origin (zero conductivity) along the semi-circle to the x-axis (large conductivity). A typical signal measured is shown by the open circles. At point [A], the surface is charged at 1.5 K. As the sample cell is cooled down, the trajectory moves along a semi-circle curve toward higher conductivity side. This behavior is interpreted in terms of the scattering of SSE by helium vapor atoms and ripplons [9]. When the sample cell temperature is stabilized at the working temperature, the trajectory stops moving (point [B]). When H atoms are introduced, the signal starts to move along the x-axis toward the origin. If the amount of H is large enough, the signal eventually disappears. This behavior means that the capacitance between the Corbino electrode and the surface charge decreases whereas the conductivity remains high.

We attribute the above behavior to the electron capture reaction which gives rise to the number decay of

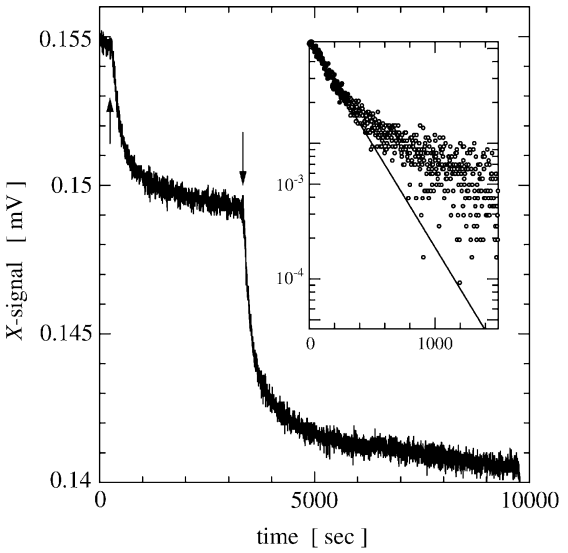

Fig. 2. Decay of SSE signal amplitude due to H atoms. The conductivity signal, which is supposed to be proportional to the number of the SSE, starts to decrease from the time point indicated by arrows when H atoms were supplied into the cell. The inset is the semi-log plot of the signal, which shows that the decay is exponential and the time constant τ is determined from the slope. This data was taken at $T = 0.2$ K.

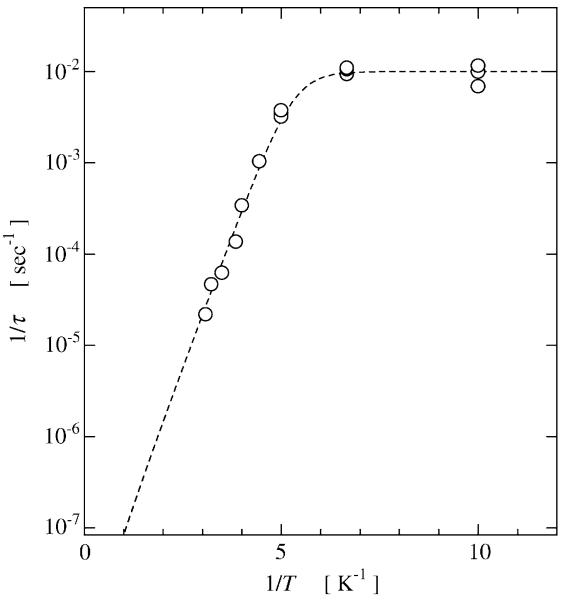

Fig. 3. Arrhenius plot of the decay rate of SSE signal, $1/\tau$. The dashed curve is obtained from the fit to the model described in the text.

SSE. We measured the decay rate of SSE after the injection of small amount of H. Fig. 2 is a typical decay curve. The x-signal of the lock-in output is plotted against time. When H atoms are introduced, as pointed by the arrows, the decay starts. By assuming that the x-signal is proportional to the total amount of SSE, we determine the time constant τ from the initial slope of the semi-log plot (the inset of Fig. 2).

The $1/\tau$ is plotted as a function of inverse temperature in Fig. 3, where the open circles represent the measured points. The lower the temperature, the faster the reaction is and it saturates below 0.2 K ($1/T > 5\,\mathrm{K}^{-1}$). The data can be fitted to the model described in the next section, which is shown by the dashed curve in Fig. 3.

3. Discussions

The model for analysis is the following. First, we will make assumptions: The number decreases of both H and e^- are only induced by electron capture reaction. The recombination of H, $H + H \rightarrow H_2$, is much slower and can be ignored. The reaction occurs only on the surface. According to these assumptions, we obtain the coupled rate equation for the total number of H (N_H) and e^- (N_e),

$$\frac{dN_H}{dt} = \frac{dN_e}{dt} = -KA_c\sigma_H\sigma_e, \tag{1}$$

where K is the reaction rate constant, A_c is the surface area of the Corbino electrode, σ_H and σ_e are the surface density of H and e^-, respectively. Since the SSE exist only on the Corbino electrode, the reaction occurs in the area of A_c. It should be noted that the loss rates of H and e^- are equal. Since H atoms in the 3D gas phase are in equilibrium with the adsorbed surface state, the surface and the volume density (n_H) of H are related by the adsorption isotherm,

$$\sigma_H = n_H\lambda_{th}e^{\varepsilon_a/k_BT}, \tag{2}$$

where $\lambda_{th} = (2\pi\hbar^2/mk_BT)^{1/2}$ is the thermal de Broglie wavelength with atomic hydrogen mass m, ε_a is the adsorption energy of H on liquid ^{4}He surface and T is the temperature. From Eqs. (1) and (2) with $N_H = Vn_H + A\sigma_H$, where A is the whole surface area of the

sample cell and V is the cell volume, the rate equation for H becomes

$$\frac{dN_H}{dt} = -\frac{1}{\tau}N_H \quad (3)$$

with

$$\frac{1}{\tau} = \frac{KN_e}{(V/\lambda_{th}e^{\varepsilon_a/k_BT}) + A}. \quad (4)$$

Since the supplied H is much smaller in amount compared to N_e, we can consider N_e in Eq. (4) independent of time and it is equal to the initial value determined by the pressing electric field. In this work, $N_e(0) = 1.2 \times 10^8$ electrons. We neglect the temperature dependence of K, which is supposed to be weak compared with the exponential dependence in the denominator in Eq. (4). Therefore, the solution of the rate equation for e^- is

$$N_e(t) = N_e(0) - N_H(0)(1 - e^{-t/\tau}). \quad (5)$$

We adjusted K and ε_a so as to fit the data and obtained $K = 4.0 \times 10^{-10}$ cm^2/s and $\varepsilon_a/k_B = 2.5$ K. The dashed curve in Fig. 3 is calculated from Eq. (4) using these values. Our model well explains the measured points. We conclude that the chemical reaction takes place on the surface. However, the measured ε_a is significantly larger than the well established value, $\varepsilon_a/k_B = 1.0$ K for H on pure ^{4}He surface [10]. Because our surface is charged with electrons, the interaction of H with the surface may be modified.

We postpone quantitative discussions until future experiment is done because the assumption that the signal amplitude is proportional to N_e is not well established. We are planning to employ the vibrating capacitor electrometer technique, the signal of which is proportional to the surface charge. Application of a strong magnetic field must be interesting. When the electron spin is aligned, the reaction rate is expected to be suppressed because the bound state of H^- is spin singlet.

4. Summary

The 2D mixture of H and e^- on the surface of superfluid ^{4}He was studied experimentally. As H atoms are introduced, the number of e^- started to decrease. From the analysis of the temperature dependence of the decay rate, we conclude that the chemical reaction of H and e^- takes place on the adsorbed surface phase.

Acknowledgements

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