Creation of a high concentration of atomic hydrogen in impurity-helium solids.

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Abstract

The exchange tunneling reactions $D+H_2 \rightarrow HD+H$ and $D + HD \rightarrow D_2+H$ were used to generate high concentrations of atomic hydrogen in impurity-helium solids. The dependence of atom concentration on the content of hydrogen in the injected gas mixture gave a maximum concentration of $7.5 \cdot 10^{17}$ cm^{-3} hydrogen atoms for an initial gas ratio H_2:D_2:He = 1:4:100.

Key words:
impurity-helium solid; atomic hydrogen; tunneling reactions; matrix isolation; quantum solids

1. Introduction

Quantum fluids and solids have been studied intensively by low temperature physicists for almost a century. We define quantum fluids and solids by comparing the interparticle spacing of the identical particles comprising the fluid or solid with the thermal de Broglie wave length of these entities. When these lengths become comparable, we are in the quantum regime. These investigations have led to many remarkable phenomena including superfluidity, superconductivity, Bose Einstein condensation in atomic gases, quantized vortices, magnetic phase transitions and a multitude of other effects. We have been investigating impurity-helium (Im-He) solids [1], which are porous complex gel-like substances consisting of individual impurity atoms and/or molecules or clusters of impurity atoms and/or molecules, each surrounded by a thin layer of solid helium. Van der Waals forces are responsible for binding together these constituents [2]. The samples are prepared by injecting a beam of mixed helium and impurity gas into a beaker of superfluid helium at $T = 1.5$ K. Snowflake like particles of the Im-He solid form in the liquid helium and fall to the bottom of the beaker where they congeal to form our sample.

We have investigated the formation and evolution of Im-He solids containing atomic hydrogen, deuterium, and mixtures of molecular hydrogen and deuterium in an attempt to maximize the hydrogen atom concentration, with the ultimate goal of creating a new quantum solid. The experiments were performed at $T = 1.35$ K.

2. Discussion

Since solid hydrogen tends to float on the surface of liquid helium, formation by the above method of an Im-He solid containing only hydrogen as an impurity was not possible. The existence of exchange tunneling reactions $D+H_2 \rightarrow H + HD$ and $D + HD \rightarrow H+D_2$ provided an alternative strategy for creating H atoms in our Im-He sample [3]. Figure 1 illustrates the time evolution of an Im-He sample prepared from an inital mixture of H_2, D_2 and He gases in the ratio 1:20:420.

The gases were mixed and then passed through a radio frequency discharge to dissociate the molecules.

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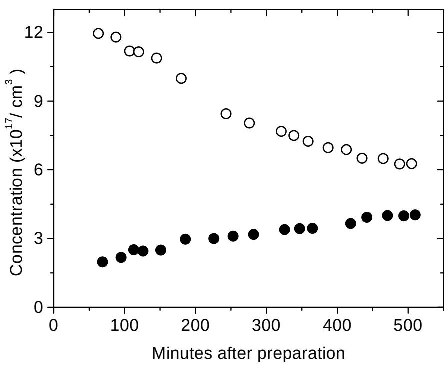

Fig. 1. Time evolution of the average concentration of H atoms (closed circles) and D atoms (open circles) after formation of an Im-He solid with an initial gas mixture ratio H_2:D_2:He = 1:20:420. Notice the buildup in the H concentration and the decay of the D concentration, mainly attributable to exchange tunneling reactions.

The beam contained H and D atoms and H_2, HD and D_2 molecules when it collided with the superfluid helium surface. The H and D atom concentrations were monitored via the intensity of electron spin resonance signals throughout the experiment. The experimental apparatus and the H and D electron spin resonance spectra for these samples have been discussed previously [4]. Very shortly after formation the sample already contained an excess concentration of H atoms because the time constant for the first of the two reactions is very short [5]. As time progressed, both reactions were involved, and the H atom concentration increased. Since the D atoms were consumed in the chemical reactions, their concentration steadily decreased. We expected the concentrations of both H and D to eventually decrease as a result of recombination. This effect was observed for gas mixtures with larger ratios of H_2:D_2.

Recently we have studied the Im-He solids formed by a variety of initial gas mixtures of H_2, D_2 and He. The ratio of impurity gases H_2 and D_2 to helium gas in the mixtures was 1:20, but the initial ratio of H_2 gas to D_2 gas was varied as follows: H_2:D_2 = 1:20, 1:8, 1:4, 1:2 and 1:1. In our studies of the concentrations of H atoms in the solids as a function of the initial gas mixture we were able to determine the best mixture for maximizing the H atom concentration. The experimental points in Figure 2 correspond to the H atom and D atom average concentrations after a storage time of 500 minutes following initial sample preparation. The optimal gas mixture of H_2:D_2:He = 1:4:100 gave a hydrogen atom concentration of $7.5 \cdot 10^{17}\mathrm{cm}^{-3}$. For a free gas of hydrogen atoms this would correspond to the onset of quantum fluid behavior at a little more than 10 mK. It may be difficult to achieve this temperature because of heating resulting from recombination of atoms into molecules and possible ortho-para conversion of the hydrogen and deuterium molecules.

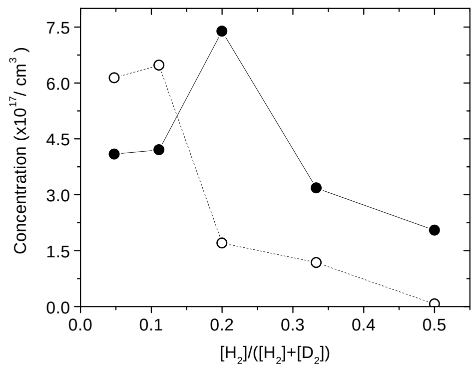

Fig. 2. The dependence of the average concentrations of H atoms (closed circles) and D atoms (open circles) in Im-He solids on the fraction of hydrogen gas in the make up gas mixture, $[H_2]/([H_2]+[D_2])$. For each point the concentrations were determined after a waiting period of 500 minutes.

We believe the majority of the H atoms in our sample tend to be closely associated with H_2, HD and D_2 molecules [6]. This is reasonable in view of the fact that the H atoms are formed via chemical reactions in the sample involving these molecules. Therefore the local concentrations of H atoms may in fact be much higher than the average concentrations. If this were the case, the onset of quantum solid effects would occur at a much higher temperature.

Acknowledgements

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