Water-Helium Condensate (Watergel) in Liquid Helium

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Abstract

The water-helium condensate is formed in He II cooled below 1.5 K as a result of condensation of the gas flow of ^{4}He with the water impurity at the surface of superfluid liquid. The experimental results indicate that the total content of water in bulk of the gel samples is less than 10^{20} molecules/cm^3 and that their density is a few percents higher than the density of surrounding liquid only. The heating of the gel samples (compact icebergs) in liquid helium inhibits its rearrangement to a more stable structure (an amorphous ice apparently) at T_d = 2.5 K when the vapor pressure P over the surface of normal liquid He I is equal to 0.2 atm, and it increases to T_d=4 K when P = 1 atm. In He gas atmosphere at SVP the "dry" icebergs start to decay at T = 1.8 K.

Key words: superfluid helium, watergel, van-der-waals complexes

1. Introduction

In this paper we present the results of our studies of the properties of metastable water impurity–helium samples that form when a flow of gaseous ^{4}He containing impurity water vapor is condensed on the surface of superfluid He II. Brief reports of the observation of a new form of water — a porous impurity–helium condensate permeated with superfluid helium — and a description of the experimental setup used to prepare the water samples have been published previously [1].

It should be recalled that features of the behavior of impurity– helium condensates were first observed by Savich and Shalnikov about 60 years ago [2] where it was observed that the properties and character of the interaction between impurity particles formed in the vapor over liquid helium depend substantially on the properties of the liquid. When an air impurity was admitted into a dewar containing normal He I a fog arose in the vapor over the He I, i.e., the impurity molecules of the gas were joined into small clusters. When the liquid was cooled below T_λ the fog disappeared, i.e., the transition from normal He I to superfluid He II was accompanied by substantial changes in the size and rate of precipitation of the clusters in the vapor over the liquid as well as in the shape of the "flakes" formed in the volume of the liquid through the agglomeration of clusters. The observations [2] served as a basis for the development of modern methods of obtaining metallic nanoparticles by the evaporation of a metal in liquid helium vapor [3] and for preparing the impurity–helium condensates formed in He II by molecular and rare gases [4,5]. According to [3,5] in cold vapor over the surface of He II the majority of impurity molecules and atoms join together into clusters with an average diameter of several nm and the characteristic sizes of the pores in these condensates are distributed over a wide range from 8 to 800 nm.

As it followed from the calculations [6] even at room temperatures H_2O molecules in the saturated vapor over the liquid should join into clusters. The trapping of water clusters consisting of several molecules — linear isomeric chains of the type $(H_2O)_N$ for $N \leq 6$ or three-dimensional "cages" for $N > 6$ — by small (about 50 nm) drops of He II traveling through a vessel containing water vapor was successfully observed in the experiments [7]. The behavior of the water clusters upon the coalescence of the drops of He II was not

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investigated earlier. However, based on the results of previous studies it was reasonable to assume that individual molecules and small clusters of water should agglomerate at the vapor–superfluid He II interface into nanoparticles with dimensions of the order of several nm, and the core of a water condensate accumulating below the surface of He II should consist of the water nanoparticles surrounded by a layer of solidified helium (so named van-der-waals complexes). Strong interaction of the complexes with surrounding superfluid liquid could result in formation of a new form of condensed water - watergel.

2. Experimental results and discussion

The first series of experiments was made in a narrow cell prepared from the glass tube of inner diameter about 9 mm with the small 1 mm outlet at the bottom filled with He II and cooled below 1.5 K. The recent studies were made in a wide 30 mm cell to diminish any influence of the wall on the gel samples.

The main results of our studies are as follows:

1. At a low rate of admission of the mixture ($\Delta P \sim$ 0.2 torr) the as-prepared gel-like cloud of the water condensate beneath the surface of the He II is transformed in a time into a denser immovable iceberg suspended on the walls of the cell. The shape of an iceberg remains unchanged at a constant temperature $T < 1.6$ K. This behavior is characteristic for gels formed in the interaction of a finely disperse suspension with a surrounding liquid matrix — a dispersion medium (sol–gel reaction).

2. As the temperature of the He II is increased above 1.6 K, the visible volume of the iceberg decreases monotonically, i.e., the structure of the sample can change smoothly as the temperature of the liquid is raised. One of the reasons for this may be the enlargement of the impurity nanoparticles and the decrease in the average size of the pores between the randomly packed particles.

3. The temperature interval in which the icebergs can exist depends strongly on the properties of the surrounding medium. In an atmosphere of gaseous helium at SVP the icebergs decompose upon heating above 1.8 K, while in liquid He I at a pressure $P = 1$ atm they can exist to 4 K. In contrast to the water condensate, impurity condensates of rare and molecular gases can exist up to 7 K. 4. The total water content in the volume of the icebergs is $\leq 10^{20}$ H_2O molecules/cm^3. The density of the icebergs is about a few percent higher than the density of the surrounding liquid. The decomposition of the water condensate in a gaseous atmosphere is accompanied by the escape of a jet of cold helium and creation of the ice flakes at the bottom of the cell or granules at the walls of the glass cell. The properties of the flakes and granules do not change upon thermocycling in the interval 1.4–5 K. Consequently, the structure of the initial condensate (watergel) and the ice particles (amorphous ice apparently) is qualitatively different.

Using the results of studies of the properties of metallic clusters in dense helium vapor [3] and of the impurity– helium samples of molecular and atomic gases [4,5], it is natural to assume that the water condensate consists mainly of water nanoclusters with an average diameter of the order of 5 nm (i.e., one cluster can contain of the order of 10^3 isomeric chains and/or three-dimensional "cages" of H_2O molecules) [7] surrounded by a layer of solidified helium. The interaction of these van-der-waals complexes with each other and with superfluid He II gives rise to a metastable porous condensate (quantum gel ?), the properties of which vary noticeably with changes in the properties of the surrounding medium.

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