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The filament formation by impurities embedding into superfluid helium

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The hydrogen molecules embedded in superfluid helium as a gas jet are shown to form long thin filaments. These filaments survived under helium transition to a normal phase demonstrating their conjugated entity. The concentration of an impurity in the core of vortex may be the mechanism of the impurity coalescence providing a cotton-like structure of a condensate obtained by the impurities contained gas helium jet introduction to He-II.

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Since the author's first study [1] a number of investigations have been devoted to introducing guest particles – atoms and molecules – from a gas into superfluid helium. It seemed obvious until the pioneer paper [2] recently appeared that the guest atoms and molecules partially already in a gas and then in a liquid stick together forming clusters determined by X-ray diffraction to have a characteristic size of about 6 nm [3]. The literature left no doubt that the geometrical shape of such clusters, at least for the atoms and simple non-polar molecules such as hydrogen and nitrogen, is grains and snowballs, as is common for any liquid and gas. The observed enhanced stability of chemically active atoms in a condensate has been attributed to either core isolation by helium [4] or primary stabilization of atoms near the clusters surface [5] but not to the filament-like structure of the condensate. In work [2] the small hydrogen grains have been preliminarily grown and then suspended in normal liquid helium. Under λ-point transition by liquid ^{4}He cooling these particles formed rather short (3 mm) filaments. These filaments aligned in a liquid under cryostat rotation along the rotation axis just as quantized vortices did, the thread density was proportional to angular velocity according to the Feynman rule. It was proved in such a way that impurity particles, hydrogen first of all, could be captured in quantized vortices in liquid helium forming prolonged structures. The capability of vortices to capture impurity particles was predicted long ago [6, 7], the simulations made in [8] showed that heavy atoms and especially molecules should hold a quantized vortex so strongly (4–8 K) that it provides the stability of a vortex inside of a rather small ^{4}He droplet. However, with the exclusion of ^{3}He and ions there was no direct experimental evidence for the vortex pinning by an impurity and no suggestion has been made about vortex pinning by an ensemble of atoms or molecules [9]. That is why the experimental evidence that quantized vortices with a core size less than one nm can trap features of at least micron size (because they have been optically observed) and survive [2] was so unexpected and intriguing.

To make the capture of an impurity in the vortex easier we decided to embed the hydrogen directly into superfluid helium using a mixture significantly diluted by helium gas; the powerful gas jet provided a short time of mixture transport to the gas-liquid interface. We considered that if the hydrogen clusters were concentrated mainly in the vortices their coalescence should occur much faster due to high local density of hydrogen in the vortex core.

We used a technique similar to that applied in [1] to introduce the impurities into superfluid helium by gas helium jet containing the traces of particles of interest. The observation method used to show the structure of a condensate just formed in a liquid was based on schlieren photography [10]. If so far the main goal was to use impurities for visualization of vortices [2], our intention was to study whether a vortex could govern the shape of an impurity cluster grown there. In particular we should like to determine either real long filament may be formed in superfluid helium or that structure would be the ensemble of small grains aligned in a chain by a vortex – such a question did not put in [2].

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The superfluid was achieved by pumping the bath of ^{4}He Dewar equipped with two pairs of optical windows. The inner diameter of the Dewar was 160 mm and the distance between the inner windows was 220 mm. The liquid helium temperature was measured by a Ge resistance thermometer immersed in liquid and calibrated against the ^{4}He saturated vapor pressure. A mixture of hydrogen and deuterium was preliminarily prepared in a cylinder at a pressure of 3–6 bars and it was allowed with using an electromagnetic valve to put into the inlet capillary at pulses of 80 msec duration with a repetition rate 6 Hz, 30-40 bursts in each injection series. The molecular source was a Dewar tube with a central capillary of 1 mm inner diameter and a nozzle of about 300 μm at its bottom. The nozzle was provided with a heater and was thermally isolated from helium vapor by Stycast 2850. The distance between the nozzle and the surface of the superfluid helium was 3–7 cm. A diagram of the cryostat and optical system is shown in Fig.1. The impurity particles in the superfluid were monitored by a shadowgraph or a schlieren technique and the images were taken by the CCD camera. The spatial resolution of the particle registration system was 20–30 μm and depth resolution was about 3 cm.

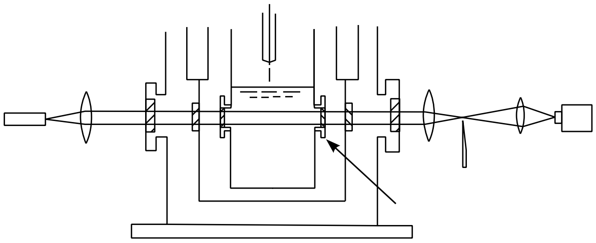

Fig.1. Schematic diagram of the cryostat. There are four optical windows two of which are shown. The arrow indicates the windows in the ^{4}He bath which extends into the vacuum space by 3 cm

Since the nearly equimolecular mixture of hydrogen and deuterium formed particles which levitated in the liquid helium, the preliminary prepared gas mixture H_2:D_2:He =1:1:200 was mainly used in our experiments, though some repetitions were performed using H_2:He= =1:100 and D_2:He =1:100 mixtures. With some delay after the series of pulses of H_2:D_2 mixture injection was terminated the numerous very long hairs, often being much longer than the window size, appeared throughout the superfluid; this was proven by changing of the focusing area of the observation technique. Such filaments were observed for the H_2:He mixture as well, though they moved randomly but predominantly upward to the surface. In the case of the H_2:D_2 mixture the motion was predominantly in a horizontal direction without any sign of upflow. Sometimes the filaments were rather short, only a few mm long and practically linear. They had a predominantly vertical position and in the case of the H_2:He mixture moved up with a velocity of 0.5 cm per second. The number of hairs slowly decreased with time and in 20 min none were observed in the field of view.

Most convenient was the observation of such filaments pinned to protuberances on the gas-liquid interface near the wall against the window's tube where the sediment usually collected as a belt (that place is shown in Fig.1 by arrow). In this case they remained in view for a long time. Some examples of such filaments are shown in Fig.2. The pictures were taken by the shadowgraph technique and the background was subtracted to

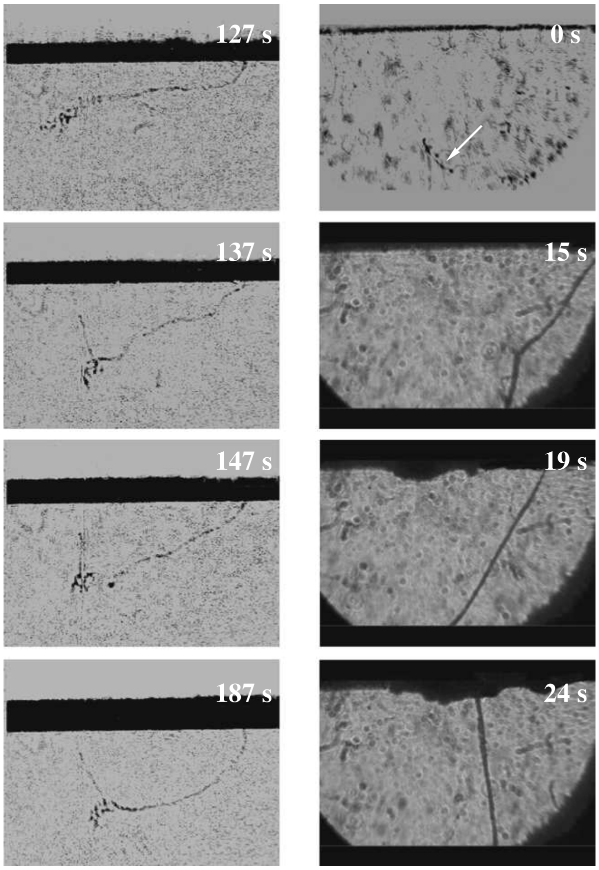

Fig.2. The examples of the filaments. The left column shows pictures of the filament produced in the superfluid phase ($T = 1.6$ K). The right column shows pictures in the normal phase. At top is the filament which survived at transition to the normal phase. The lower pictures are a thicker thread which was not pinned at the interface.The number in each picture shows the time (in sec) elapsed after the jet was terminated, for the experiment shown in right column the jet was contained only pure helium in order to induce fast HeII – HeI transition by heating

get better contrast. The pictures in the left column display the filament behavior in the superfluid phase. The thick black horizontal line upward is the gas-liquid interface. Numbers in the picture indicate the elapsed time after the first jet shot. The length of filament presented there was around 3 cm and its diameter, nicely constant along the whole thread, appeared to be sufficiently less than 50 microns. We could observe the filament swinging in the flow of the normal component, its upper end was pinned to the protuberance, and the lower end was probably out of focus. It was clearly obvious that the feature was levitated in the liquid helium, i.e. it was consisted of hydrogen-deuterium mixture.

We never observed the filaments in experiments with normal liquid helium, even when the temperature was rather close to the λ-point; only small randomly moving in a liquid spherical particles were seen in the case. It is interesting to note that the aftercooling below 2.1 K did not cause the filaments to appear in He-II. However, when created below the λ-point, they survived the subsequent transition from superfluid to normal state; an example is seen in the top picture of the right column (the arrow indicates the filament) in Fig.2. That observation seems to us rather symptomatic because it proves that the formed filament is not a simple collection of separate hydrogen grains kept together but a conjugated and stable entity made of non-polar hydrogen. A thicker thread of about 100 μm diameter shown in pictures 2,3 and 4 was formed later, probably by a number of filaments merging into the fiber. Its behavior was different from that of the filaments in He-II because, as seen in Fig.2, it was not pinned to a particular place.

To confirm whether the filament formation is the main channel of hydrogen coalescence or is an occasional though very interesting event, it makes sense to compare the number of hydrogen and deuterium molecules in the filament with that injected into the cryostat. Estimating the hair diameter as 30 microns and assuming its particle density equal to that in solid hydrogen, i.e. $3{\cdot}10^{22}$ per cm^3, one can estimate the number of molecules in a 3 cm long hair as $N_v = 1 \cdot 10^{18}$. The total number of molecules entering the cryostat during one injection calculated simply from the gas mixture consumption is $2.5 \cdot 10^{20}$. The probability for hydrogen to be captured in liquid helium that we measured earlier in a similar situation of intensive counterflow of evaporating helium is about 0.2 [9]. The volume of liquid helium accessible for optical observation by the schlieren technique, taking into account its depth resolution of 3 cm and window diameter of 3 cm, consists of only 1% of its total volume. Thus, in our observation field we should see an average of $N_{av} = 0.5 \times 10^{18}$ molecules. The N_v and N_{av} values turn out to be rather close to each other and as a rule we really have found one hair in-focus. This means that in the frame of our technique the condensation of guest particles into filaments is possibly the main process of coalescence but is at least quite effective. In particular, the solid condensate in superfluid helium including that studied in [11, 12] should remind cotton rather than snow.

Such a non-polar substance as hydrogen could form at the sedimentation in uniform media only grains (in volume) or films (at the surface), some special other factors are necessary to induce the appearance of hydrogen-deuterium filaments. Because this feature grew only in the superfluid phase one should attribute the effect to the main properties of superfluid ^{4}He, namely to the absence of hydrodynamic friction, to the super – heat conductivity by the counter flow, and to the existence of quantized vortices which are produced by any outer violation. The gas jet is such a violation because the dimple in the liquid helium interface at the place of the jet entry into liquid was 3–4 mm deep and had a 5 mm diameter (see Fig.4b in [13]).

Unfortunately, we cannot prove at present whether the mechanism of filament formation under impurity embedding as molecules or nanoclusters directly into HeII is different from that realized in [2] and consisted in already formed grains alignment along a vortex. However our visual observations indicate in favor of the mechanism consisted in mutual interlacing of very thin wires formed inside vortices as nuclei into threads, this process possibly can take place in normal liquid He as well.

Provided the filaments are the main shape of condensate particles below λ point, the total rate of impurity sedimentation in the superfluid phase should be much faster than in normal liquid. Indeed, because every collision of molecules and clusters in liquid helium leads to their coalescence, the rate of condensation is proportional to surface-to-volume ratio in sediment, the last being more for elongated structures. In addition, the hooking of very long filaments, resulting eventually in their interlacing, proceeds especially fast.

The visual observation has shown that the thickness of hydrogen-deuterium filament is practically the same along the entire thread as if some maximal diameter of the hydrogen filament exists. Of course, the levitating mixtures of hydrogen with deuterium or neon are unique from the viewpoint of steady pattern creation. But for understanding the structure of heavier impurity-helium solids it makes sense to analyze what may happen in the case of embedding particles other than hydrogen and deuterium. For small clusters containing less than 10^6 atoms or molecules gravity has no role [5] and their behavior should be the same as for hydrogen; the changes in the energy of molecule-vortex interaction for different molecules are not so large [8]. Large clusters of pure hydrogen would be moved up by Archimedean force and, as we have seen, retain their shape. Large heavy impurity clusters should sink down to the bottom of a cell (with a pinned vortex or without it) and as we believe with retaining their shape as well. This problem is very interesting from a theoretical point of view but it looks even more intriguing for applied science because of the possible synthesis of micro and even nanowires; such materials are of great significance for microelectronics and catalytic chemistry, and the high price of a desirable product could justify using the superfluid helium. It is very interesting as well to carry out a similar study for high pressure helium just below the pressure of its solidification. The variations of vortex core size are noticeable [14] but mainly the solidification of helium in the first and second helium layers around the core should have large impact. Such investigations are now in progress.

We should stress at the conclusion that in our experiments on direct imbedding of hydrogen and deuterium into superfluid helium as in the experiments described in [2] the size of filaments is many orders of magnitude larger than the vortex core size.

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