testifies to the formation of a one-dimensional conductor with a substantially higher conduction electron concentration than on the remaining surface.

An experiment aimed at measuring the quantity $(dI/dZ)/I$ gives the idea of how narrow the region of an electronic spectrum singularity near the boundary is. This measurement is realized by applying to the STM piezo-electric cell, which displaces the tip in the Z direction, an alternating voltage with a frequency that lies above the feedback band, and by measuring the variable signal amplitude as a function of the surface X, Y-coordinates. In the quasiclassical tunneling model, the current is an exponential function of Z, with the exponent proportional to $W^{1/2}$ (W is the work function). Consequently, in the above experiment, the work function versus surface coordinates is measured.

As is evident from Fig. 6, variations in the exponent are rather high. In this case, immediately at the boundary there is an anomaly only about 0.25 nm in half-width, which is even somewhat less than the interatomic distance. Therefore, at the boundary there forms a minimal-width conductor — one atom in width.

The experiments outlined above furnished extensive data on the structure of peculiarities on the surface of a cleaved facet, which reflects on the atomic scale the processes occurring in the rupture of a metal, and on their specific electronic properties.

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Impurity nanocluster structures in liquid helium

L P Mezhov-Deglin

1. Introduction

The first experiments aimed at the preparation and study of the properties of impurity nanocluster condensates in superfluid helium-II were pioneered by A I Shal'nikov and staged in Chernogolovka in the early 1970s [1–3]. Shal'nikov's original idea of attempting to prepare a film of atomic (metallic) hydrogen by the condensation of H atoms on a substrate cooled to a temperature T on the order of 1 K looked quite attractive. It was well known, for instance, that thin bismuth films deposited in a vacuum on a substrate cooled to $T \approx 4.2$ K are superconductors, although bismuth under ordinary conditions makes up a semimetal with a low density of free charge carriers. However, these films are metastable and recrystallize on substrate heating above 20 K, so that on being subsequently cooled the film resembling a massive sample behaves like a semimetal. Is it possible to prepare a quasi-two-dimensional film of atomic hydrogen by depositing H atoms on a solid substrate cooled below 1 K? How is it possible to localize free hydrogen atoms on the substrate surface and minimize the probability of their subbarrier tunneling accompanied by $H + H \to H_2$ recombination of atoms that find themselves at the neighboring sites? Will such a film of atomic hydrogen possess metallic conduction and all the more superconduction? The answers to such questions could be provided only by a direct experiment which would be hard to stage even nowadays. As we are aware today [4], recombination of atomic hydrogen restricts the maximum content of free H atoms in three-dimensional deuterium – hydrogen condensate samples impregnated with superfluid HeII, which are prepared by condensing a gas beam that contains hydrogen and deuterium atoms on a superfluid liquid surface. The content is limited by the values of $n_H \sim 7 \times 10^{17}$ cm^{-3}, so that the relative content of H atoms in the molecular $H_2/HD/D_2$ matrix, which forms the gel frame (solid skeleton of the gel), does not exceed 1%.

Shal'nikov discussed the possibility of arranging the research work in this area, which lies at the interface between low-temperature chemistry and physics, with staff members of several Moscow institutes, in his native Leningrad Fiztekh, and lastly in the Institute of Solid State Physics (ISSP) in Chernogolovka, to whose high-capacity low-temperature base formation he had largely contributed. It was agreed that the low-temperature facility would be assembled at the ISSP, while the high-temperature atomic hydrogen source ('dissociator') for it would be possible to make at Fiztekh. However, the initial work plan had to be radically changed with time. We are reminded that in the early 1960s, during the formation period of the scientific center of the USSR Academy of Sciences in Chernogolovka, research into the methods of accumulation and storage of free radicals, including the atoms of molecular gases in molecular matrices in the simplest case, were on the list of the main research areas of the future center. Interest in this research was stimulated by the fact that its findings in case of success might be used, for instance, to increase the specific momentum of the fuel for

rocket and aircraft engines, because the energy released in the recombination of hydrogen or nitrogen atoms is comparable to the energy liberated in the burning of hydrogen in oxygen. But in the mid-1960s, interest in this subject waned rapidly. One of the reasons lay with the appearance of a monograph [5] published by the US National Bureau of Standards. On the basis of experiments and theoretical calculations performed at the NBS, it arrived at the conclusion that films with a thickness of about ten molecular layers or more, prepared by deposition of atoms and molecules of the substance under investigation on a substrate cooled to $\sim 4\,\mathrm{K}$, could contain no more than tenths of a percent of a free radical (and hundredths of a percent for H atoms) owing to the thermal instability of such samples and that further lowering of the substrate temperature was wholly immaterial. These estimates were one to two orders of magnitude below the requirements of the industry. The realization of the idea Shal'nikov had come up with called for rather high consumption of resources and human energy. And it comes as no surprise that it met not only with support, but also with serious opponents whose reasoning relied on the conclusions of the monograph [5].

Apart from a group of ISSP staff members who had wide operational experience in the area of low-temperature physics, for the pursuance of the investigations described in Refs [1 – 3] we succeeded in recruiting the staff members of the Chernogolovka Branch of the Institute of Chemical Physics, who specialized in the area of the physics and chemistry of gas beams. As revealed by our analysis, one of the main causes for developing the instability of a film prepared by high-vacuum deposition of the atoms and molecules of a substance under investigation on a cold substrate is that the kinetic energy of atoms produced in the dissociator during thermal dissociation of molecules or, for instance, under dissociation induced by microwave radiation exceeds 10^3 K, i.e., exceeds the substrate temperature by more than two orders of magnitude. This fact substantially determines the kinetics of chemical reactions on the surface and is responsible for the thermal instability of thick films with a high content of free radicals.

An efficient way of lowering the kinetic energy of the atoms emanating from a dissociator consists in the use of a dense cold buffer gas, for instance, helium-4 which can hardly enter into chemical reactions with the substance under investigation and is easy to cool down to several kelvins in experiments with liquid helium. Moreover, to ensure a high rate of heat removal from the film surface and prevent the development of thermal instability due to atomic recombination, the substrate can be precovered with a layer of superfluid HeII. One drawback of this method is evident: if the rate of the gas flow condensing on the HeII surface is low, the impurity atoms and molecules can manage to coagulate in clusters in the cold helium vapor above the liquid, so that the flow reaching the vapor – superfluid liquid interface will mostly contain the clusters of the impurity being investigated. However, this may equally be considered as a virtue of the technique proposed. It has been known that a neutral impurity particle at temperatures on the order of 1 K in superfluid HeII and in the saturated vapor above the liquid surface is, as a rule, surrounded by one to two atomic layers of solidified ^{4}He, i.e., the so-called van der Waals complex forms, which consists of the impurity nanoparticle and the layer of solidified helium around it. This lowers the likelihood of impurity nanoparticle coalescence with crystallite forma-

tion in the bulk of impurity condensate which forms on the surface and in the bulk of HeII owing to the mutual attraction of the complexes [6, 7]. This also opens up the possibility of studying the low- and ultralow-temperature kinetics of impurity nanocluster formation in the gas phase and the tunnel chemical reactions in individual clusters and in impurity-helium nanocluster condensates (impurity gels) containing free radicals.

In the first experiments [1 – 3], we studied the feasibility of stabilizing nitrogen atoms in superfluid helium. Nitrogen atoms were obtained by letting gaseous ^{4}He with an admixture of molecular nitrogen N_2 flow through a gas-discharge tube. Nitrogen was not adopted at random: the recombination of nitrogen atoms is accompanied with luminescence in the visible range, which matters when conducting a pilot study. Judging by the estimates given in Refs [2 – 4, 6, 7] and in the subsequent publications of the Cornell and Finnish groups [8 – 15], the relative concentration of nitrogen atoms in the frame of the impurity gel being formed in the liquid, i.e., the $[N]/([N] + [N_2])$ ratio, may range into dozens of percent. Therefore, going over from the conventional technique of the vacuum deposition of molecular-gas atoms on a cold solid substrate to the introduction of a gas jet into superfluid HeII cooled below 1.6 K has led to an increase in the relative free-atom concentration in the bulk of nitrogen condensate by nearly two orders of magnitude in comparison with the estimates of the maximum attainable free-radical concentration in the molecular matrix, given in the monograph [5]. Subsequent to nitrogen, a series of pilot experiments was carried out in the same facility with admixtures of oxygen, deuterium, and hydrogen.

Despite the obvious turn-around of the subject of investigation, Shal'nikov continued to take a keen interest in the progress of our work. His remarks and advice were especially valuable as he himself possessed wide experience in thin-film preparation by condensation of material on a cold substrate [16], elaborated techniques of producing colloids and gels [17], and in 1946 published a paper [18] jointly with P Savich, which reported the observation of the coagulation peculiarities of gaseous impurities (most of all, air) admixed to a glass Dewar vessel with liquid helium cooled below 2.5 K. It was found that cooling the liquid below the normal HeI-to-superfluid HeII transition point T_λ is accompanied by a qualitative change in coagulation mechanisms of impurity particles produced in the vapor above the liquid: from a finely divided suspension or fog, which fills the entire Dewar vessel for $T > T_\lambda$, to large flakes slowly settling in the liquid upon cooling it below T_λ. When the air was replaced with hydrogen, the reverse picture was observed, as well — the disintegration of large impurity flakes floating on the liquid surface into small particles upon raising the fluid temperature above T_λ. The disintegration was accompanied by the formation of an 'aureole' above a large hydrogen particle. These observations suggested that the structure and properties of impurity particles produced in the dense helium vapor above the liquid surface, as well as the properties of impurity condensate samples formed in liquid helium, depend crucially not only on impurity composition, but also on the properties of liquid helium.

At present, investigations of impurity-helium condensates are being pursued by experimental groups both at the ISSP, [19 – 24] and at the Chernogolovka Branch of the Institute of Energy Problems of Chemical Physics [6 – 15, 25] forming the parts of the RAS scientific center in Chernogolovka, as well as

at other laboratories, in particular, at Cornell University (USA) [4, 8–14] and Jyvaskyla University (Finland) [15, 25]. Approaching completion is the preparation for joint [ISSP RAS, Chernogolovka; Joint Institute for Nuclear Research (JINR), Dubna; Institute Laue–Langevin (ILL), Grenoble (France)] neutron structure investigations of the gels of deuterium and heavy water in HeII, employing the ILL neutron beam [23, 24, 26, 27].

2. Preparation technique of impurity gel samples

Figure 1a displays the schematic diagram of the facility which was first described in Refs [1–3], presently employed in the preparation of impurity condensates containing atoms of a molecular gas. The overall concentration of gaseous impurities in the jet of gaseous ^{4}He, which is prepassed through a gas-discharge tube (a microwave dissociator mounted in the upper part of the device) cooled by liquid nitrogen, does not exceed several percent. The relative content of the atoms of the molecular impurity under investigation in the gas jet at the dissociator output, for instance, the $[N]/([N] + [N_2])$ ratio in experiments with atomic nitrogen, amounts to $\sim 90\%$. Since the specific density of solid hydrogen is almost two times lower than the density of liquid helium, to stabilize hydrogen atoms in the samples of impurity condensate in the bulk of HeII requires introducing, along with hydrogen, deuterium or other heavier impurities (for instance, rare gases) into the gas jet.

The diameter of the outlet at the bottom of the tube is equal to ~ 0.5 mm. The outgoing gas jet is immediately directed to the superfluid liquid surface, which fills a glass cup mounted inside a helium Dewar vessel directly under the outlet of the discharge tube. A thermomechanical pump is employed to fill the cup with superfluid HeII. The fluid temperature is $T \approx 1.5$ K. The gas pressure at the dissociator inlet is several torrs higher than the saturated ^{4}He vapor pressure in the cryostat volume. The minimal spacing between

the dissociator outlet and the HeII level in the cup amounts to $h = 2$ cm. The shape and color of the gas jet depend greatly on this spacing. For $h \leqslant 3$ cm, the jet takes on the shape of a needle (Fig. 2a), its point abutting against the cone-shaped meniscus formed on the surface of HeII. In experiments with nitrogen, the jet itself and the nitrogen condensate, which slowly settles to the bottom of the cup in the superfluid liquid bulk, glow in green [1–3, 6, 15]. For a larger spacing ($h > 3$ cm), an orange downward-broadening plume is observed in lieu of the needle, i.e., the recombination glow of excited ^{4}He is prevalent in a warmer jet [11, 25].

The nitrogen condensate glow in the HeII bulk decays within only several dozen seconds after impurity feed cessation and resumes when the liquid warms up above T_λ (Fig. 2c). The recombination glow in the gas jet can be separated from the condensate glow observed during sample preparation by modulating the output power of the microwave generator, which is connected to the gas-discharge tube of the dissociator, with a frequency of several hertzes. This leads to the modulation of the gas jet glow (Fig. 2b). The green glows of the gas jet and the condensate arise primarily from the $N + N \rightarrow N_2$ recombination of nitrogen atoms and the relaxation of excited nitrogen molecules in the nitrogen matrix, so that in line with modern observational data [6, 8, 15], impurity nanoclusters, which contain the atoms and molecules of the impurity being introduced, are produced even in the jet above the liquid surface. As indicated by subsequent investigations, the samples of impurity-helium condensates formed in HeII may exist not only in superfluid HeII, but in the normal HeI liquid, as well (Fig. 2c). The disintegration temperature for the samples extracted from liquid helium depends greatly on the helium-4 vapor pressure. At atmospheric pressure in the Dewar vessel, for instance, the impurity condensates of molecular gases decay rapidly (sublimate) on heating above 7 K.

The above preparation technique of impurity-helium samples is employed in experiments staged to investigate gel
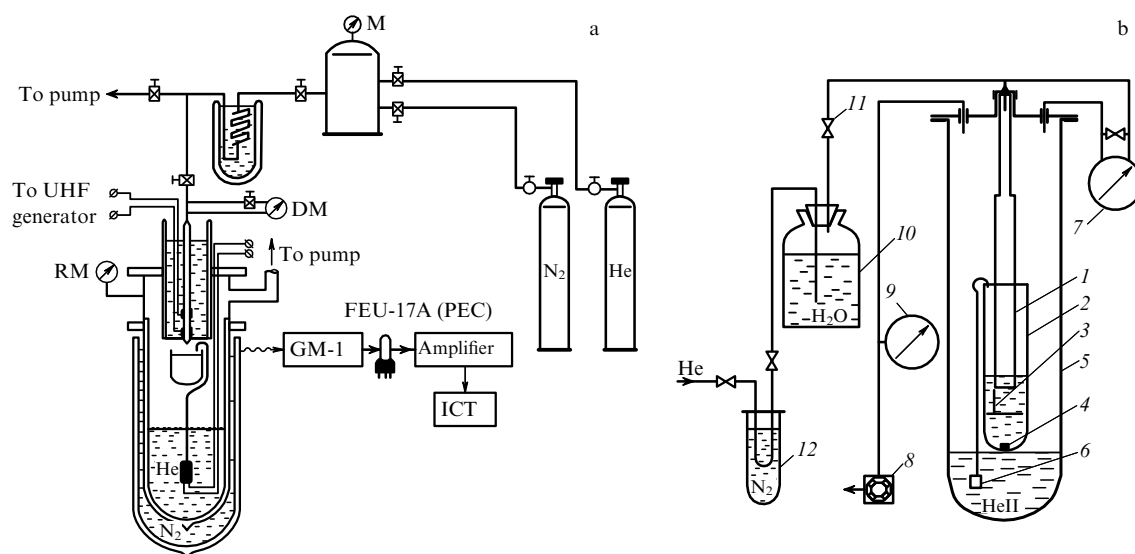

Figure 1. Preparation of impurity-helium nanocluster condensates: (a) schematic of the facility with a microwave dissociator intended for operation with molecular and atomic gas impurities [2]: M — manometer, DM — differential manometer, RM — reference manometer, GM — grating monochromator, and UHF — ultrahigh-frequency generator; (b) schematic of the facility employed for work with the vapors of liquids [21]: *1* — mixture intake tube, *2* — quartz ampoule, *3* — teflon disk 30 mm in diameter with two resistive thermometers mounted on it, *4* — heater, *5* — helium Dewar vessel, *6* — thermomechanical pump employed for filling the ampoule with HeII, *7* — differential manometer, *8* — mechanical pump, *9* — manometer, *10* — water-containing vessel, *11* — valve for feeding the gas mixture, and *12* — nitrogen-cooled trap.
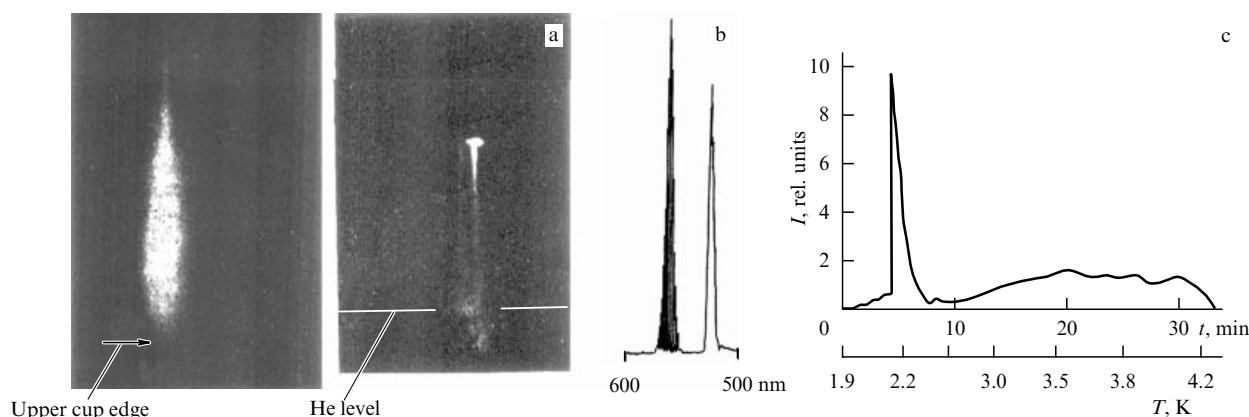
Figure 2. (a) Evolution of the shape of a gas jet containing nitrogen atoms and molecules, when raising HeII level in the upper cup (Fig. 1a): left frame — the spacing between the dissociator outlet and the HeII level is about 4 cm; right frame — the spacing is reduced to 3 cm; the liquid temperature amounts to ~ 1.7 K, the pressure in the dissociator is ~ 10 Torr higher than the helium vapor pressure in the Dewar vessel, and the initial $He - N_2$ gas mixture composition is close to 100 : 1. (b) Glow spectrum of the nitrogen – helium mixture in the Dewar vessel during sample preparation: one can see the lines (modulated) of the gas jet spectrum and the continuum of the condensate glow in the HeII bulk. (c) Afterglow of the condensate containing nitrogen atoms, when raising the temperature of liquid helium (luminescence at a wavelength of ~ 523 nm).

structure by X-ray diffractometry and ultrasonic attenuation, in studies of tunnel chemical reactions in free-radical-bearing samples by EPR techniques, and in optical investigations into the mechanisms of clusters and nanocluster condensate production as well as of the relaxation of excited helium molecules in HeII. Among the disadvantages of this technique are the difficulties associated with controlling the processes occurring during condensate production and accumulation (the directed gas jet strongly mixes the liquid and the condensate produced in the cup), as well as great impurity losses in the course of sample preparation, because a substantial part of the gas flow introduced into the Dewar vessel is carried away with the helium vapor into the outward pump line. Furthermore, to prevent the dissociator outlet from being plugged up, advantage can be taken here of only those impurities whose condensation temperature is known to be below the boiling temperature of liquid nitrogen.

Figure 1b demonstrates the schematic of the facility which we designed to prepare and study the properties of impurity gels of materials that solidify at temperatures well above the boiling temperature of liquid nitrogen [20 – 22], for instance, the gels of water (a polar liquid) or ethanol (a weakly polar liquid). Unlike the facility depicted in Fig. 1a, this facility permits us to follow the course of the preparation of the samples, their evolution, and their decay with an increase in surrounding fluid temperature or with the lowering of the HeII level, when the samples are embedded in cold helium vapor. To keep watch on the formation of the samples and their subsequent evolution, advantage is taken of a video camera (not shown in the diagram) mounted outside of the device. The camera is connected to a video recorder and a computer, which facilitates further information processing. These observational data are discussed in Section 3.2 (see Figs 5 and 6).

In the preparation of a water gel, gaseous ^4He is first passed through a vessel *10* with water. Next, the gas mixture saturated with water vapor arrives at the input of a glass intake tube *1*, whose internal diameter was varied from 9 to 25 mm in different experiments. In the 'wide experimental cell' shown in Fig. 1b, the lower edge of tube *1* is submerged in superfluid HeII cooled to the temperature $T \sim 1.4$ K.

As shown by our operational experience, the optimal preparation conditions for the samples of impurity-helium water condensate are as follows: HeII temperature $T \leqslant 1.5$ K, and excess pressure $\Delta P \leqslant 0.6$ Torr in the gas jet at the inlet to tube *1*, i.e., an order of magnitude lower than in the facility diagrammed in Fig. 1a. In this regime, the flow rate of the gas mixture delivered to the inlet tube from the external system measures $\leqslant 5$ cm^3 s^{-1} under normal conditions, with all entering ^4He condensing on the HeII surface in the working cell. The equilibrium water vapor density in the $^4He + H_2O$ mixture is $\sim 3\%$ at room temperature, so that less than 6×10^{21} H_2O molecules enter the working cell during the mixture intake time $t \approx 20$ min. An appreciable part of these molecules is deposited on the walls in the upper part of the intake tube. One can visually discern two to three icy belts several centimeters wide, formed in the upper part of the tube. These belts supposedly correspond to different crystal ice structures (it is well known that ice possesses a hexagonal structure at the triple point; several phase transitions are observed on further temperature lowering [28], while in the evaporation of water molecules on a substrate cooled to 10 K a layer of amorphous ice forms). Therefore, the water content in the water gel samples formed in the HeII bulk does not exceed 10^{21} molecules cm^{-3}. It is noteworthy that the lower part of the tube remains transparent over a distance of several centimeters above the helium-II surface in the optimal sample preparation regime. However, upon lowering the HeII level one can see the formation of an opaque ice deposit on the tube walls at the site where the interphase boundary was previously located. This deposit forms due to the decay of impurity-helium condensate in the helium vapor. The dynamics of the effects observable at the vapor – liquid interface for high mixture intake rates will be discussed below in Section 3.2.

3. Main results of an investigation

3.1 Structure and properties of the gels formed by molecular and rare-gas impurities in HeII
3.1.1 Structural investigations.
To estimate the typical dimensions of impurity clusters, use was made of X-ray diffractometry techniques [8 – 11]. Experiments on investigating the
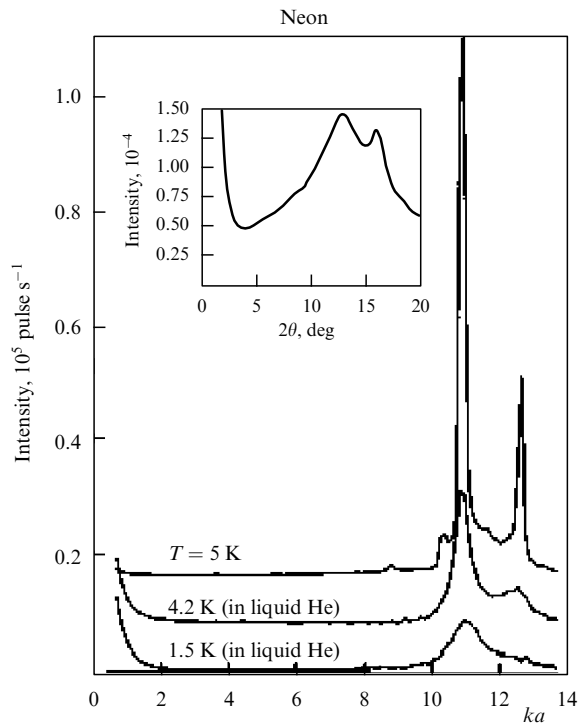
Figure 3. X-ray scattering from a neon sample submerged in liquid helium. The inset shows the whole spectrum. The background corresponding to the scattering by liquid helium has been subtracted from the lower plots. Here, a is the lattice constant of solid neon, and k is the transferred momentum. The peak location is determined by the lattice constant, and the peak width by cluster dimensions. According to the estimates of the authors of Refs [8, 10], the characteristic cluster dimension $d_{mean} \sim 6$ nm (similar estimates for a deuterium sample yield $d_{mean} \sim 9$ nm).

Figure 4. Ultrasonic wave damping in the volume of an experimental cell: open circles — pure HeII, closed and open squares — nitrogen gel; measurements at frequencies of 3.16 and 5.33 MHz; T_1 — the temperature below which the main contribution to the sound damping is made by the superfluid helium bulk; T_2 — the temperature at which the damping curve flattens out. In the view of the authors of Ref. [9], the increase in ultrasonic wave damping in the temperature range between T_1 and T_2 is attributable to the decrease in viscous penetration depth $d_{visc} = (2\eta/\omega\rho_n)^{1/2}$. The plateau for $T > T_2$ corresponds to sound propagation primarily via large channels in the gel. In this sample, the characteristic pore dimensions, which are estimated from the values of d_{visc} for T_1 and T_2, are varied through a range $8 - 800$ nm.

X-ray scattering from particles formed by rare- or molecular (D_2, Ne, N_2, Kr) gas impurities in a dense helium vapor above the HeII surface, as well as the scattering from gel samples formed from them in the superfluid liquid bulk, were carried out on the synchrotron source (X-ray diffractometer) of the Brookhaven National Laboratory, USA. The γ-quantum energy was equal to 17.3 keV in experiments with neon, and to 8 keV in experiments with other samples. One of the results of this research — the angular dependence of scattered intensity with a sample of the neon gel in liquid helium for different sample temperatures — is depicted in Fig. 3. The peak location is determined by the lattice constant a, and the peak width by the cluster dimension d. According to the authors' estimates, the typical dimension of the neon clusters is $d_{mean} \sim 6$ nm; similar estimates, for instance, for a deuterium sample yield $d_{mean} \sim 9$ nm. It is noteworthy that the scatter in dimensions of the particles that find their way to the liquid surface is rather wide: from separate atoms or molecules to clusters whose diameter exceeds the above typical dimension d_{mean} by more than an order of magnitude. In principle, it is also possible to make a judgement about the typical pore dimensions in the nanocondensate from the data on inelastic X-ray scattering by a sample of impurity-helium condensate in HeII. The estimates given in Refs [8, 11] suggest that the pore dimensions in the neon sample ranged between 8 and $\geqslant 400$ nm.

More detailed data about the effective nanocondensate pore dimensions and the evolution of the internal structure of freshly prepared samples with an increase in HeII tempera-
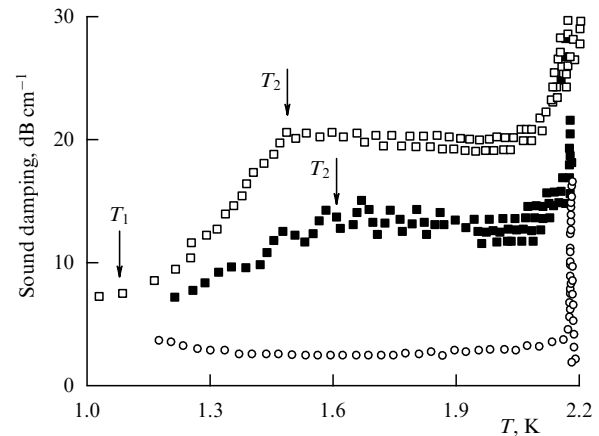
ture were obtained in experiments involving ultrasonic wave transmission through a cell filled with the condensate under investigation. The technique of ultrasonic wave damping measurements in D_2, Ne, N_2, or Kr gel samples is discussed in detail elsewhere [9]. It was found that the velocity of sound in gels is close to the velocity of sound in pure HeII, but the ultrasonic wave damping in the gels is orders of magnitude stronger than in the liquid and increases rapidly with temperature. Figure 4 portrays the ultrasonic wave damping measurement data for a sample of nitrogen gel in a bulk of liquid helium at two frequencies. The open circles correspond to pure helium; the closed and open squares to helium with the $N_2 - $He condensate; the measurements were performed at frequencies of 3.16 and 5.33 MHz. Here, T_1 is the temperature below which the main contribution to the sound damping is made by the superfluid helium bulk, and T_2 is the temperature at which the damping curve flattens out.

The authors attribute the increase in sound damping over the temperature range between T_1 and T_2 to the shortening of viscous penetration depth $d_{visc} = (2\eta/\omega\rho_n)^{1/2}$. Here, η is the viscosity, ω is the sound vibration frequency, and ρ_n is the normal component density. The plateau for $T > T_2$ corresponds to sound propagation primarily through large channels in the gel. The characteristic pore dimensions in this sample, estimated from the values of d_{visc} for T_1 and T_2, are varied through a range $8 - 800$ nm. Upon raising the fluid temperature above T_λ and subsequent cooling, the sound damping in different gel samples increases significantly. This may be attributed to an irreversible increase in nanocluster aggregate dimensions and an increase in the average density of the samples with the rise of the temperature.

These findings are consistent with observations of glow induced by the free-N-atoms-bearing nitrogen condensate with an increase in the fluid temperature (Fig. 2c), as well as with a smooth decrease, observed in our work [22], in apparent dimensions of the samples of water or ethanol

gels — icebergs floating below the liquid surface — with an increase in the HeII temperature above 1.6 K. All these changes are attributable to the aggregation of small-sized clusters, controlled by the thermally activated diffusion of molecules or atoms across the cluster surface.

3.1.2 Investigations of atomic diffusion and tunnel chemical reactions in impurity-helium condensates in HeII by EPR techniques. The properties of the samples of impurity gels containing nitrogen, hydrogen, and deuterium atoms were studied by electron paramagnetic resonance (EPR) techniques in a cylindrical quartz cell at frequencies on the order of 9 GHz (see Refs [10 – 14] and references cited therein). The power scattered in the resonator did not exceed several microwatts. The absorption of the microwave radiation by N, H, or D atoms was registered in samples of various compositions. The content of atoms in a sample under investigation and variations in their relative concentrations with time or with increasing fluid temperature were numerically estimated by comparing the EPR spectra of the samples being investigated and standard test samples in the same resonator. The studies of nitrogen gel properties demonstrated the possibility of preparing samples containing up to 10^{21} N_2 molecules in 1 cm^3 of the sample bulk. In this case, the *local number density* of nitrogen atoms in the molecular frame amounted to 10% (on the order of 4×10^{20} atoms cm^{-3}).

The most interesting results were obtained in the studies of hydrogen and deuterium atomic diffusion and of tunnel chemical reactions in the samples prepared by condensation of dissociated gas mixtures containing hydrogen and deuterium atoms and molecules in superfluid helium at temperatures $T \sim 1.35 - 1.5$ K [4, 11 – 14]. The initial impurity content $[H_2]:[D_2]:[He]$ in the beam was varied over wide limits from $1:20:400$ to $1:1:100$ in different experiments. The average number density of impurity molecules in deuterium – hydrogen gels was about the same, viz., on the order of 10^{20} cm^{-3}. However, the average concentration of H and D atoms in freshly prepared samples depends heavily on the initial mixture composition and may vary with time or with an increase in sample temperature. For instance, in a sample prepared from a mixture of optimal composition $1:4:100$ at $T = 1.5$ K and then cooled to $T = 1.35$ K, the content of hydrogen atoms *monotonically rose* with time (almost two-fold in 500 min) to attain a value of $(7.5 \pm 3.0) \times 10^{17}$ cm^{-3}. In this case, the content of deuterium atoms in the initial sample was several times higher than that of hydrogen and *lowered* by a factor of $1.5 - 2$ with time (the number density of deuterium atoms in this sample was $\sim 1.5 \times 10^{17}$ cm^{-3}). Long-term observations of H and D atomic concentration variation in one of the optimally composed samples with a high concentration of atomic hydrogen ($\sim 5 \times 10^{17}$ cm^{-3}) at $T = 1.35$ K [14] showed that, during the subsequent 40 h, the number density of hydrogen atoms *hardly changed* and the number density of deuterium atoms *lowered* by ~ 2.5 orders of magnitude in comparison with the initial concentration of about 10^{17} cm^{-3}.

The increase in atomic hydrogen content and the simultaneous concentration lowering of atomic deuterium in the samples of optimal composition is an indication that the tunnel exchange reactions

$$D + H_2 \rightarrow HD + H \quad \text{and} \quad D + HD \rightarrow D_2 + H$$
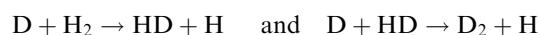
dominate in these samples.

At the same time, the concentrations of H and D atoms monotonically *decrease* with time in a sample prepared from an $[H_2]:[D_2]:[He]$ mixture with a $1:2:60$ component ratio, where the initial density of H atoms is three times higher than in the previous mixture and exceeds the density of D atoms, i.e., the increase in atomic hydrogen content due to exchange reactions is balanced out by the recombination of atoms that found their way to the neighboring sites of the solid frame of the deuterium – hydrogen gel:

$$H + H \rightarrow H_2 \quad \text{and} \quad H + D \rightarrow HD.$$
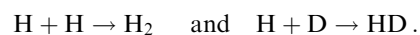
The hydrogen atoms migrate over the sample due to the exchange reactions

$$H + H_2 \rightarrow H_2 + H \quad \text{and} \quad H + HD \rightarrow HD + H,$$

while the migration of deuterium atoms is controlled by the exchange reaction

$$D + D_2 \rightarrow D_2 + D,$$
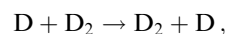
which proceeds with a rate four orders of magnitude *lower* than that of the hydrogen exchange reaction. That is why the mobility of free hydrogen atoms in deuterium – hydrogen gels is far greater than the mobility of deuterium atoms.

Observations of satellite lines in the EPR spectra of microwave absorption by H and D atoms suggest that these atoms are stabilized in clusters which make up the impurity condensates of the mixtures of H_2, D_2, and HD molecules. In particular, every H atom is surrounded by a layer of D_2 molecules, which impedes the hydrogen atom recombination. A hydrogen atom surrounded by a shell of D_2 molecules *becomes stable* because it can neither migrate through the shell nor interact with the D_2 molecules via the $H + D_2 \rightarrow HD + D$ exchange reaction, since this reaction is *endothermic*.

By taking advantage of the results of these measurements and the data of previous X-ray structural investigations, Bernard et al. [14] estimated that the *local* concentration of H atoms *inside the* $HD-D_2$ *clusters* can be as high as $\sim 10^{20}$ cm^{-3}, i.e., the relative concentration of in-cluster hydrogen atoms can be on the order of 1%. Hence, it follows that a cluster involving about 10^4 D_2 and HD molecules may also contain about 10^2 H atoms, and the average separation of the hydrogen atoms is equal to ~ 2 nm. This gives hope that it will be possible to discover cooperative quantum effects upon cooling the deuterium – hydrogen gel with a high content of H atoms to a temperature on the order of 100 mK.

It is noteworthy that the characteristic cluster dimensions in the samples prepared from dissociated pure deuterium were equal to 9 ± 3 nm, the volume content of D_2 molecules amounted to $7 \times 10^{20} - 3 \times 10^{21}$ cm^{-3}, the content of D atoms in the gel to about 1×10^{18} cm^{-3}, so that the *local in-cluster* content of deuterium atoms amounted to 2×10^{19} cm^{-3}.

3.1.3 Optical glow investigations of the condensates of molecular gases. The experimental cell employed to study the emission of a gas jet containing nitrogen atoms, as well as the emission of nitrogen – helium condensate in the HeII bulk at temperatures $T \sim 1.5 - 1.7$ K, was described in detail in Ref. [15]. To distinguish between the processes occurring in the gas jet and in the condensed phase, the authors modulated

the power of the high-frequency generator that fed the dissociator (the duration of the HF pulses was on the order of 200 μs, and the pulse repetition rate ran to 500 Hz). The work involved the registering of atomic nitrogen glow in the green spectral region ($\sim 520 - 525$ nm), as well as of the emission in the blue spectral region ($\sim 240 - 360$ nm) arising from excited nitrogen molecules produced upon atomic recombination. Both of these transitions are forbidden in the gas phase, and therefore observations of this kind may serve as a test in studies of impurity cluster production in a gas jet directly above the liquid surface and of impurity-helium condensate formation on the surface and in the bulk of the fluid. It was observed that the most intense emission arises from the crater region. The crater emerges on the liquid surface under the action of the gas jet which travels with a velocity of ~ 70 m s^{-1}. This clearly demonstrates that the impurity aggregation processes, which are accompanied with cluster production and the recombination of nitrogen atoms in the cluster bulk, most actively proceed in the dense helium vapor above the liquid surface. In the bulk of the fluid, the radiation intensity falls off rapidly with depth at a distance of several millimeters from the crater surface. The molecular emission spectra arising from the gas jet, the crater surface, and the liquid bulk appear similar and are shifted to the red side in comparison with the well-known emission spectrum of single molecules in the gas phase. In exactly the same way, the emission spectra of atomic nitrogen from the jet above the liquid surface, the crater surface, and the liquid bulk turned out to be similar. And so in all cases a significant part is played by the processes occurring inside impurity clusters in the gas jet or in the nanocondensate produced in the coalescence of nanoparticles in the liquid bulk, and not by the radiation of separate isolated atoms or molecules. From the known rate of gas jet travel it may be estimated that about 10^3 N$_2$ molecules are contained in the volume of the clusters that penetrate into the liquid bulk, so that the characteristic cluster dimensions are equal to ~ 3 nm. These estimates are consistent with the above data to an order of magnitude.

As an extension of these investigations, the next work [25] was concerned with a study of the fluorescence associated with the relaxation of metastable helium atoms and excimer helium molecules produced when a jet of pure helium-4 was passed through the dissociator. Also studied in the work was luminescence quenching upon addition of a hydrogen impurity to the helium jet. The measurements were made in the $580 - 720$ nm interval. Excited He atoms in the triplet state and excimer molecules were found to accumulate in the crater region on the HeII surface. The observations of radiation generation in the liquid bulk may be related to the reactions proceeding on the liquid surface — recombination of excited atoms and excimer molecules accompanied by the emergence of helium ions and electrons on the surface. The subsequent electron – ion recombination gives rise to highly excited atomic states and excimer molecular states on the crater surface and in the liquid bulk. The luminescence quenching upon addition of the hydrogen impurity to the gas jet is attributable to efficient energy transfer from helium to the H$_2$ molecules.

3.2 Water and ethanol gels in HeII
Water constitutes the basis for organic life in the Universe, and it comes as no surprise that detection of water traces in any form on distant planets immediately attracts the attention of scientists who specialize in various fields. We became interested in the possibility that there may exist a new form of water — an impurity-helium water condensate (a water gel) at low temperatures. As indicated by our experiments [19 – 24], introducing gaseous ^{4}He with an admixture of water vapor into a vessel with superfluid HeII does give rise to a semitransparent condensate at the vapor – liquid interface. The condensate comprises a water gel which exists only at the liquid-helium temperatures. The water gel structure has yet to be elucidated, although by analogy with the condensates of molecular gases it may be assumed that the frame of this gel is formed by the (H$_2$O)$_n$ type nanoclusters surrounded by one to two layers of solidified ^{4}He. This assumption is supported by the findings of Nauta and Miller [29] who reported the observation of water nanoclusters (from dimers to hexamers) inside helium droplets about 50 nm in diameter, which were flying through a vessel filled with water vapor. The facility employed to prepare water and ethanol gels in superfluid HeII is outlined in Fig. 1b. When elaborating the preparation technique and elucidating the existence domain of the water and ethanol gel samples, investigations were carried out in several cells of different designs. In particular, when studying the water-gel existence domain in experiments with a 'narrow cell' [19 – 21], the lower end of the intake tube with an internal diameter of 9 mm was sealed off. Made in the bottom of the tube was a clearing hole 1.5 mm in diameter, which served the purpose of feeding superfluid helium to the working cell from the external glass test-tube.

The optimal conditions for the preparation of samples are as follows: the fluid temperature $T = 1.4$ K, the excess pressure $\Delta P \leqslant 0.6$ Torr in the gas flow at the intake tube inlet, and the gas flow rate $\leqslant 3$ cm^3 s^{-1}. The semitransparent cloud (the jelly-like condensate) being formed at the vapor – liquid interface in the tube slowly shifts along the tube walls into the HeII bulk. The intake was stopped when the cloud thickness became equal to the cell diameter. For a constant fluid temperature, the cloud transformed within a time period of ~ 10 min into an oval sample (iceberg) with an average diameter close to the cell diameter, which floated under the fluid surface. As the HeII level was smoothly lowered, it was possible to observe the iceberg shifting stepwise down (diving) to hover under the HeII surface again at the instant the upper iceberg edge turned out to be above the fluid level. As this took place, the temperature of HeII in the cell was virtually invariable; pressure jumps were noted instead, i.e., the stepwise iceberg displacement is attributable to the outflow of a cold gas jet upwards from the prominent part of the sample surface warmed by an external light source (the water condensate absorbs the visible light much more strongly than liquid helium and the glass tube walls).

By repeating the intake, it was possible to prepare several icebergs located one above the other along the cell. This allowed us to study the evolution of the shape and decomposition of the icebergs under the subsequent increase in the ambient fluid temperature, as well as to observe the decay of 'dry' icebergs (extracted from HeII) in the helium vapor at pressures above several torrs. Upon evaporation of HeII from the cell, it turned out that the temperature of 'dry' icebergs, which assemble on the bottom of the cell, monotonically rises from ~ 1.4 to 1.8 K over a rather long period ($10 - 20$ min, depending on the brightness of outside lighting). In this case, the readings of the upper and lower thermometers mounted in the cell were practically equal. Above 1.8 K, the indications of the thermometers begin to significantly diverge, and abrupt temperature jumps with an amplitude up to 0.3 K are
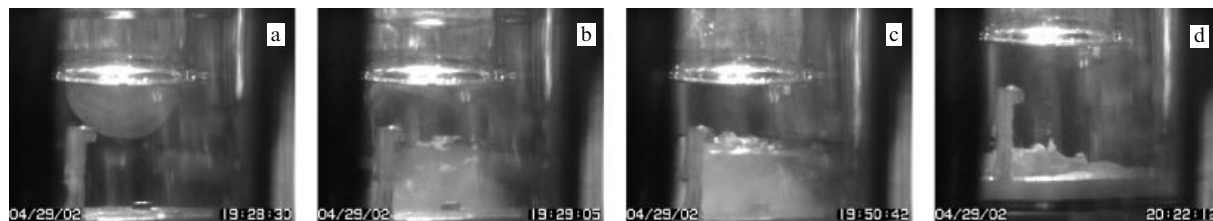
Figure 5. Water gel formation in HeII and decay of a sample with the lowering of the HeII level in the experimental cell depicted in Fig. 1b. The fluid temperature is constant ($T = 1.4$ K). In the photographs, one can see the lower edge of the mixture intake tube (*I* in Fig. 1b), the thermometer anchored on a steel needle at a distance of 1 cm above the teflon substrate, and the thermometer located on the substrate surface. Frames, from left to right, are as follows: (a) water gel cloud at the outlet of the intake tube; (b) gel layer formed at the cloud incidence on the substrate (the HeII layer is above the edge of tube *I*); (c) evolution of the sample shape with the lowering of the fluid level (the HeII level is below the upper thermometer), and, lastly, (d) water gel decay in ^{4}He vapor (the fluid level is below the substrate, the substrate temperature $T \sim 2.18$ K and rises to $T \sim 2.6$ K at 1 cm above the substrate, and the vapor pressure in the Dewar vessel is $P \sim 2.2$ Torr).

observed. The temperature jumps are accompanied with cracking of the icebergs and their decrease in volume. For T above 2.2 K, the jumps no longer occur, and the temperature of the helium vapor inside the cell rapidly rises above 4 K. The total volume of the pieces of ice, which are formed on the bottom and walls of the cell in the decomposition of the icebergs, is approximately two orders of magnitude smaller than the initial iceberg volume. Consequently, the water content in the bulk of the icebergs does not exceed several percent, so that the water concentration in the iceberg bulk does not exceed 10^{20} molecules cm^{-3}. This is consistent with the above estimate for the water content in the gas which condensed on the HeII surface in the course of mixture intake. In this experiment, the fluid temperature in the Dewar vessel is maintained constant ($T \sim 1.4$ K). Accordingly, the helium vapor pressure in the Dewar vessel is constant and amounts to $P \sim 2$ Torr, but the HeII level is located lower than the bottom of the working cell, and therefore the cell with the condensate gradually becomes warmer. When the helium vapor pressure in the Dewar vessel rises to the atmospheric pressure, the temperature of 'dry'-iceberg decay increases to about 4 K. The decay of 'dry' icebergs is accompanied by an intense outflow of gaseous ^{4}He through the nozzle in the lower part of the intake tube. Therefore, a rather large amount of liquid helium is contained inside the icebergs extracted from HeII. This liquid helium vaporizes when the condensate is heated to 1.8 K by an external light source (for comparison: according to mass spectrometric composition analysis of the gas produced in the decay of molecular impurity-helium condensates [7] extracted from the liquid, for each impurity molecule there are up to 12 He atoms, which favors the idea that impurity van der Waals complexes emerge in the dense helium vapor).

Investigations into the iceberg shape evolution and decay in liquid helium with an increase in ambient fluid temperature showed that the icebergs can also exist for temperatures higher than T_λ in the bulk of normal HeI liquid. The temperature T_p whereat gaseous helium is released from the sample and the icebergs actively decay (thaw) in normal HeI depends crucially on the vapor pressure in the device. For $P \approx 0.2$ atm, the temperature of the onset of decay is $T_p \approx 2.5$ K; it rises to $T_p \approx 4$ K with an increase in pressure to 1 atm.

For a high intake rate ($\Delta P \geqslant 1$ Torr), the initially plane liquid – vapor interface twisted within minutes after the onset of the mixture intake, and a meniscus up to 1 mm in height was clearly visible at the interface, i.e., a transparent water-gel layer about 1 mm in thickness began to grow along the HeII surface from the tube walls towards the center. When the intake was continued, at the point in time the layer covered the entire HeII surface, the pressure in the tube increased stepwise by a factor ranging into the dozens ($\Delta P \geqslant 20$ Torr), resulting in extrusion of the condensate layer and the tube-filling liquid into the outer test-tube. The water gel extrusion through a 1.5-mm outlet at the bottom of the cell was accompanied by gel decay — at the bottom of the outer test-tube there was accumulated a fine-grain powder (supposedly, amorphous ice) whose properties remained invariable upon evaporation of the fluid and subsequent temperature rise above 7 K. Similar results were obtained in a series of preliminary experiments with ethanol gel whose samples turned out to be less stable than water gel samples, as well as in experiments with deuterium gel.

In connection with preparing for the pursuance of joint experiments (ISSP, Chernogolovka; JINR, Dubna; ILL, Grenoble) aimed at studying the structure of low-capture material gels, first and foremost deuterium and heavy water gels, by neutron spectroscopy techniques [22 – 24, 26, 27], which require samples larger than 10 cm^3 in volume, the working cell diameter was increased to 3 cm. Accordingly, the diameter of the mixture intake tube was increased, as well. One version of the design of this cell is shown in Fig. 1b. The results of observing the water gel formation and decay in a wide cell are given in Fig. 5. The fluid temperature was constant ($T = 1.4$ K), so that the helium vapor pressure in the Dewar vessel ran into $P \sim 2$ Torr. In the photographs given in Fig. 5, one can see the lower edge of the mixture intake tube, the upper thermometer mounted on a steel needle at a distance of 1 cm from the surface of the teflon disk (substrate), and the lower thermometer mounted on the surface at the center of the disk. For a low mixture intake rate, on the walls of the wide tube there forms a semitransparent condensate cloud which slowly sinks to the working cell. In frame (a), one can see an oval condensate cloud at the outlet of the mixture intake tube. Seen next (b) is a condensate layer formed when the cloud descended onto the disk surface. The HeII level is located ~ 1 cm above the edge of the intake tube. The next frame (c) serves to illustrate how the shape of a sample lying on the substrate evolves with the lowering of the HeII level: the part of the iceberg protruding above the fluid surface gradually decays. In the last frame (d), where the fluid level is ~ 1 cm below the disk and the substrate temperature has risen to 2.6 K, one can see separate pieces of ice arisen by decaying the sample of the water gel. Figure 6 exemplifies the
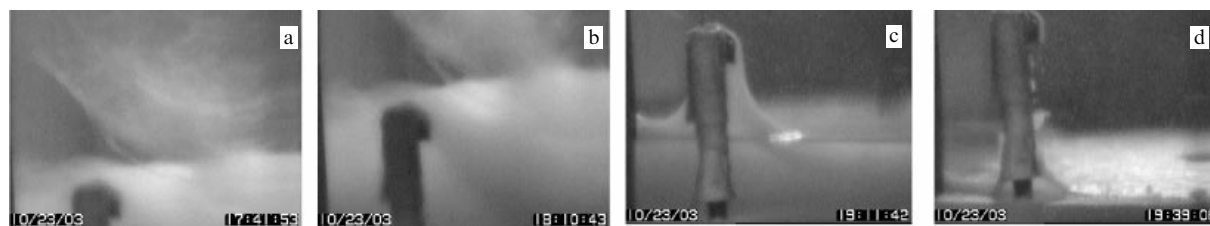
Figure 6. Deuterium gel formation in HeII and decay of the sample upon lowering the HeII level in the experimental cell shown in Fig. 1b. The fluid temperature is constant ($T = 1.4$ K). Frames a, b — formation of the sample, and c, d — decay upon lowering the fluid level.

observations of a deuterium sample formation and decay under the same conditions (the scale of the images was increased ~ 1.5-fold).

It is noteworthy that the condensate layer is observed to grow along the vapor–liquid interface for a high mixture intake rate in the wide cell just as in the narrow one. This growth is accompanied with a pressure jump and extrusion of the condensate layer from the wide tube into the outer test-tube at the point in time when the layer completely covers the fluid surface in the intake tube. That is why the new version of the cryostat, which is intended for the pursuance of joint structural investigations of deuterium and heavy water gels at the ILL neutron beam facility [24], allows for the forced extrusion of the growing condensate layer from the intake tube into the working cell in the course of sample preparation. The results of these investigations will be subsequently employed in the development of a facility intended to verify the feasibility of a new technique of ultracold neutron accumulation and storage in impurity gels in superfluid HeII cooled to a temperature on the order of several millikelvins [23, 24, 26, 27].

4. Conclusions

We list below the main results of the investigations conducted.

(1) A technique was developed for the preparation of new nanocluster materials in superfluid helium — impurity-helium condensates of molecular and atomic gases (impurity gels), which are stable at low temperatures, including samples containing atoms (free radicals) of molecular gases.

(2) The properties of impurity-helium condensates produced by condensation of molecular and atomic (inert) gases were investigated by synchrotron X-ray diffractometry, mass spectrometry, EPR, and optical and ultrasonic spectroscopy techniques. It is valid to say that the dispersive system (the frame) of impurity-helium condensate (gel) being formed in HeII comprises impurity clusters surrounded by one to two layers of solidified helium, and superfluid HeII penetrating the pores serves as the dispersive medium. The average cluster dimensions are about $5 - 10$ nm, and the characteristic pore dimensions in the sample measure $5 - 1000$ nm. The total impurity content in the bulk of a freshly prepared sample is $N \leqslant 10^{21}$ atoms or molecules per cm^{-3}, so that the average gel density exceeds the ambient fluid density by no more than a few percent. The local number density of nitrogen atoms in the molecular frame of nitrogen N_2 gel amounts to 4×10^{20} cm^{-3}, while the local number density of hydrogen atoms in the frame of deuterium–hydrogen $D_2 - HD$ gel may be as high as 10^{20} cm^{-3}.

(3) A technique was elaborated for the preparation of impurity-helium condensates of substances whose transition temperature to the liquid state is much higher than the boiling temperature of liquid nitrogen, and investigations were made of the properties of water and ethanol gel samples at temperatures above 1.4 K. The domain of existence in liquid helium and in helium vapor was determined for these samples. Estimates suggest that the total density of H_2O or C_2H_5OH molecules in the bulk of a freshly prepared gel sample is less than 10^{20} cm^{-3}, so that the water and ethanol gel densities are close to the densities of gels on the basis of molecular or rare gases. A new metal cryostat intended for the study of gel structure by neutron spectroscopy techniques was subjected to a running test.

As far as we know, the immediate research tasks set at different laboratories include an in-depth study of tunnel chemical reactions in matrix-isolated condensed systems at low and ultralow temperatures, including attempts to expose cooperative quantum effects in deuterium–hydrogen condensates with a high content of hydrogen atoms, exploration of possibilities for employing impurity nanocondensates as low-temperature energy sources and catalysts, as well as studies into potential uses of heavy water and deuterium gels as efficient moderators of cold neutrons, suited for the accumulation and storage of ultracold neutrons in a vessel filled with superfluid HeII at temperatures on the order of 1 mK.

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