Experimental Report

Proposal: **3-15-72** **Council:** 4/2012

Title: Evolution of the structure of nanostructured icy samples of heavy-water and deuterium in a broad temperature range - SANS study

This proposal is a new proposal

Research Area: Physics

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Samples: D2O (heavy water); D2 (deuterium)

Instrument	Req. Days	All. Days	From	To
D22	8	3	26/10/2012	29/10/2012

Abstract:

Slow neutrons (UCN, VCN, CN) are coherently scattered by nanoparticles that provides a powerful tool for VCN reflectors, for VCN storage in traps, for "quasi-specular" CN reflectors and for cooling VCN. Our recent X-rays measurements had shown that on heating a water nanocluster-gel sample above 4 K in He gas atmosphere, one can observe formation of highly-dispersed ice samples consisting of a mixture of amorphous and cubic ice (Ic), where the inner structure of the sample changes from amorphous to cubic Ic and then to commonly observed hexagonal ice Ih with increasing the temperature from 80 to 200 K. This observation openes up unique opportunities for improving reflecting properties of nano-structured reflectors by replacing diamond nanoparticles (with hardly-removable hydrogen contaminations) to nanoparticles from low-absorbing materials like pure D2 or D20 at reasonably high temperatures. We propose to perform a set of SANS experiments, using our specialized optical cryostat for in-situ preparation of the gel samples, for studies of nanostructured ice samples in the temperature range from 1.6 to 25 K (for deuterium ice) and from 1.6 to 200 K for heavy water ice samples.

We studied experimentally the following points previewed in our proposal 3-15-72:

a) Variations in the size distribution of impurity nanoclusters in dispersion systems (backbone, or skeleton) of heavy water and deuterium gel samples in superfluid helium He-II when heating samples to temperatures close to the critical temperature in He-II also when heating samples in bulk of normal helium He-I up to the boiling temperature of T=4.22 K at the normal pressure of P=1 atm.

b) Study the range of existence of gels in the bulk of liquid helium and in He gas atmosphere in a wide temperature range below and above $T_{boiling}$.

c) Evolution of the structure of D_2O and D_2 high dispersed icy samples following the decay of the gel in He gas atmosphere when heating the deuterium icy samples up to ~ 20 K, and up to ~ 200 K for the heavy water icy samples.

In addition to previous measurements at the D22 instrument with neutrons with the wavelength of ~6.1 A, now we expand the neutron wavelength range; the neutron transmission and scattering were measured with neutrons with the wavelengths of 6, 12, 18 and 24 A (two last values are comparable with the dimensions of nanoclusters in the backbone of the gel samples). Moreover we develop also the technique of observations: the beam transmission and scattering were measured at three different heights along the gel sample; at the distances of ~ 10, 20 and 30 mm above the bottom of the experimental cell (positions 1, 2 and 3) accordingly. Thus we could compare evolution of properties of the samples of different density of impurity nanoparticles when changing external parameters. Due to certain restrictions in the beam time allocated for this experiment (requested 8 days, allocated 3 days plus 2 days thanks to kind support of the D22 responsibles added in the last moment) we could study properties of only two deuterium gel samples and one heavy water gel sample only in the temperature range of up to 200K. Treatment of raw experimental data (more than 500 runs) will demand a lot of time and will be continued later in the Institute of Solid State Physics RAS in Chernogolovka. Our preliminary results are the following:

1) We confirm long-term stability of the structure of impurity gel samples cooled below the temperature of 1.8 K. Increase in the temperature of surrounding superfluid He-II from 1.8 K to 2.15 K (i.e. 0.03 K below the critical point for He-II in the bath) resulted in significant increase of the fraction of nanoclusters with small sizes of ~1-2 nm in the gel backbone comparable with the wavelengths of cold neutrons used. One reason for this transformation could be related to a drastic decrease in the thermal conductivity of the gel dispersion system (liquid helium in nanopores connected with each other by narrow channels), thus to poorer thermal stability of the system. It is well known that for liquid helium in pores of aerogel or gelsill samples with the pore sizes of ~10nm, or smaller, one can observe transition from superfluid He-II with extremely high thermal conductivity to BEC droplets the localized in pores and connected via narrow channels (no far order, no super-fluidity) at temperatures far smaller than the critical temperature in bulk liquid.

2) From results of measurements of the beam transmission and scattering in the gel samples in He-II in the cell of 28 mm inner diameter, one can conclude on observations of multiple scattering of long wavelength neutrons.

3) Basing on measurements of beam transmission and scattering in the gel samples at temperatures near to and above 4.2 K one can conclude on observations of the decay of the gel samples when heating them in atmosphere of He gas at the pressure of P = 1 atm (at the temperature of T ~ 4.5 K for D_2 gel and at the temperature of T~ 20 K for D_2O gel). We are going to compare results of our recent SANS measurements with deuterium and heavy water gel samples with the results of our X-ray studies of phase transitions in icy samples of conventional water produced on decay of watergel, where we observed transitions from amorphous ice to BCC nanocrystalline structure with the mean size of ~6- 8 nm at the temperature T~120 K and then from BCC to conventional HCP ice nanocrystals with the mean sizes of nanoparticles equal to ~ 80 nm at the temperatures of ~150 K and ~200 nm at the temperature of 200 K. In principle, D_2 or D_2O particles of such sizes could form an excellent reflector for VCN and UCN, but we should check the time stability of such the samples at different temperatures. As it was mention in our proposal our investigation is a part of the broader ILL Programme of investigations of the nanoparticle reflectors.