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Nucleation and crystal growth of ^{4}He in aerogel

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Outline

Crystallization of liquid in bulk state is a well-understood phenomenon. But crystallization in a restricted geometry is not well studied. Frost heaving seen in cold climates is a complex phenomenon with freezing of water in soil, which could produce large enough force to deform the stratum. Also in engineering, it relates to a number of applications such as freezing method,food freezing excavation. However, since it is not easy to eliminate the influence of non-uniformity of the medium and the impurities in such a complicated system, it is difficult to perform reproducible experiments. The crystallization in an ordinary medium is so complicated as to treat many processes such as transport of viscous flow and latent heat. Although it is well known that static properties such as elevation of melting pressure in a restricted space changes, basic physics for the dynamic properties is not understood very well. In order to approach such a problem, we investigate crystallization of ^{4}He in aerogel as a porous material.

To use ^{4}He is very beneficial on studying the crystallization in a porous material. ^{4}He remains as liquid state down to absolute zero due to its large quantum effect. The liquid has a superfluid state below 2 K, and it's viscosity becomes very small. By performing experiments with superfluid ^{4}He at very low temperature, it is possible to study in a clean environment without impurities. Owing to superfluid properties, we can ignore the effect of the viscosity of liquid even in a restricted geometry and owing to the peculiar property of crystallization at very low temperature, the latent heat is negligible in the study of crystallization. The quantum effects in the crystallization also can be expected. Superfluidity is a manifestation of quantum effects, ^{4}He crystals are also a quantum solid whose atoms move by the zero point vibration. The supersolidity exhibiting superfluidity is a major interest in the study of ^{4}He crystals. Quantum effects including supersolidity may appear as a new phenomenon in the dynamics of crystallization in restricted geometry.

Aerogel is a porous material having a fractal structure in the nano-scale region. Aerogel's porosity is from 90 to 99.5 % and its density is very low. Due to its high transparency, we are able to observe clearly crystallization in the porous material by direct visualization.

As a method for obtaining an overpressure above the melting pressure, the blocked capillary method is well known. It is the method to prepare solid ^{4}He by cooling of the high pressure liquid, let it hit the melting curve and change the whole liquid to solid by further cooling, and finally the solid under the pressure higher than 25.3 bar. However, it is impossible to investigate temperature dependence of crystallization in low temperature region by this method. So we took a method of obtaining the overpressure by mechanical pressurization of the sample cell by using Pomeranchuk idea. Pomeranchuk cell enables us to control the pressure of the sample cell under constant temperature and investigate temperature dependence of crystallization in low temperature region.

Previous research of crystallization in aerogel by visualization revealed the dynamical phase transition of the growth mode. It grows via avalanche in low temperature region and via creep of the interface in high temperature region. Measurement of crystallization rate indicated crystallization proceeds by macroscopic quantum tunneling at low temperatures, and, by thermal activation type at high temperatures.

In this thesis work, we investigated temperature dependence of nucleation processes and the ^{4}He crystallization in aerogel by cooling.

Nucleation process was directly investigated by measuring the nucleation probabilities. Critical overpressures at which the first ^{4}He crystal appeared during pressurization were measured 50 times at each temperature. The temperature dependence of the mean critical overpressure confirmed quantum nucleation in low temperature region and thermal nucleation in high temperature region. Crossover temperature was consistent with the result of crystallization rate measurement, which approximately coincided with the dynamic transition temperature of the crystal growth mode.

Meanwhile, it happened to occur in a 96% porosity specific aerogel that liquid remains in some parts of the aerogel even though other parts of the aerogel were filled with crystals in high temperature creep region. Once such a situation is formed, the application of additional pressure does not further crystallize the liquid. Here we call this left uncrystallized liquid as "liquid pocket". This "liquid pocket", however, began to crystallize via avalanche when the system is cooled below a particular temperature. The crystallization of the liquid pocket in lower temperature suggests the possibility of the mass transport mechanism through the solid which surround the liquid pocket , since solid density is larger than liquid one. Crystallization of the liquid pocket by cooling raise intriguing issue of the mass transport through crystals associated with supersolidity in aerogel.

Also by changing systematically the pressure and temperature, we attempted to elucidate the mechanism of crystallization by cooling. We intentionally left the liquid in aerogel, and cooled down the system. Crystallization started at a certain temperature by cooling not only with liquid pocket in aerogel but also with crystal partially filled or no crystal in aerogel. The temperature at which crystallization started by cooling depended on pressure. Furthermore, we examined the reproducibility of the crystallization temperature by cyclic cooling and warming. No hysteresis was observed in crystallization temperature among cooling and warming. It was found that the crystallization temperature could be defined sharply like a phase transition. Since mass needs to be supplied to aerogel for crystallization, we proposed the possibility that mass transport sharply occurred at a given temperature from bulk solid. Experiment of stepwise cooling provided temperature dependence of the crystallization rate, which increased toward low temperature. From these crystallization experiments, we could obtain a crystallization phase diagram in aerogel. There are three phases; liquid, solid, liquid-solid region. It also suggested that melting pressure curve in aerogel has a peak unlike the bulk melting pressure at around the dynamical phase transition temperature. We are supporting the possible mechanism of mass transport for crystallization by cooling due to superfluid transition of dislocation core.