Critical Supersaturation of ^{3}He-^{4}He Liquid Mixtures: Decay of Metastable States at Ultralow Temperatures

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We report the results of the first systematic study of the phase separation from supersaturated ^{3}He-^{4}He liquid mixtures of dilute phase in the temperature range from 130 mK down to 400 μK. We find that the degree of critical supersaturation becomes almost temperature independent below about 10 mK while it increases with temperature above about 10 mK.

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The nature of metastable states and the decay rate of such states have long been among the most interesting problems in statistical mechanics, both experimentally and theoretically [1]. Recently the nucleation phenomena at ultralow temperatures have received much interest because of the possibility of observing the crossover of the nucleation process from the classical regime, thermal fluctuation, to the quantum regime, quantum fluctuation or quantum tunneling. Quantum nucleation is a kind of macroscopic quantum phenomenon.

The system of a ^{3}He-^{4}He liquid mixture seems ideal for the study of nucleation phenomena. First, the system is extremely pure. Second, the study can be made in a wide temperature range over almost three decades. In order to cool the system down to an ultralow-temperature region, we need a heat exchanger of a large surface area. It is known, however, that the surface of materials immersed in a ^{3}He-^{4}He liquid mixture is covered with ^{4}He. So, any surface is unlikely to be effective in nucleating the ^{3}He concentrated phase [2]. This is the third advantage.

The nucleation of the ^{3}He phase from a supersaturated ^{3}He-^{4}He mixture of dilute phase was discussed by Lifshitz, Polesskii, and Khokhlov [3] in connection with the theory of quantum kinetics of phase transitions developed by Lifshitz and Kagan [4]. Lifshitz, Polesskii, and Khokhlov obtained the expression for the rate of nucleation W in metastable supersaturated mixtures as a function of temperature T and the degree of supersaturation, Δx_3, which is the deviation of ^{3}He concentration from the equilibrium value. They found a critical dependence of W on Δx_3 in both the classical and quantum regimes. That is, the index of the exponential function in W is proportional to $(\Delta x_3)^{-7/2}$ for the quantum regime and to $(\Delta x_3)^{-2}T^{-1}$ for the classical regime. They, therefore, suggested the existence of the rapid nucleation line below the binodal line. On one side the rate of formation of the critical nuclei is virtually zero while on the other side the supersaturated mixture breaks up practically instantaneously. So it may be possible to observe such critical supersaturation, $\Delta x_{3,\mathrm{cr}}$, experimentally. Furthermore, they estimated the crossover temperature and $\Delta x_{3,\mathrm{cr}}$ to be around 14 mK and about 0.15 at 0 K, respectively. In the quantum region, $\Delta x_{3,\mathrm{cr}}$ becomes temperature independent. Experimentally, the supersaturation phenomena in the dilute phase of ^{3}He-^{4}He mixtures were noticed by the Cornell group [5] and the Ohio group [6]. As far as we know, however, there has been no systematic study, especially from the viewpoint of the crossover.

The preliminary results of our experiments down to 2 mK were reported in [7,8]. In this paper we show the results down to 400 μK obtained with some modified arrangements and improved measuring techniques. The main modification is that we have two concentration gauges.

It is known that decomposition experiments are severely influenced by heterogeneous nucleation starting from inhomogeneities or from the walls. In the present case wall effects may be negligible, as mentioned in the beginning. In the dilute phase, a temperature inhomogeneity easily produces a concentration inhomogeneity due to the osmotic pressure effect (heat flush effect). So the process of realizing the supersaturated state should be done under the condition of constant temperature and slowly enough so as not to produce flow of ^{3}He. We apply the pressure sweep method with a superleak, through which only the zero-entropy superfluid component of ^{4}He flows into the cell to pressurize the sample mixture, or out to depressurize it. The method does not produce any appreciable flow motion of ^{3}He if the whole liquid is in the dilute phase and the process is performed slowly enough [9]. According to the arguments by Lifshitz, Polesskii, and Khokhlov, one may reach the rapid nucleation line almost without regard for the process speed as long as a good homogeneity is attained. We have two capacitance-type concentration gauges of large gap distance (0.5 and 0.2 mm). They are separated by about 120 mm in order to check the homogeneity of the ^{3}He concentration and to see whether the decomposition takes place simultaneously in the whole liquid. We have three thermometers in the cell to check the temperature homogeneity. The consistency of the concentration and the temperature homogeneities are checked with the relation of the osmotic pressure equilibrium condition.

The sample cell is schematically shown in Fig. 1. It is fastened to a copper nuclear stage. The heat exchanger is

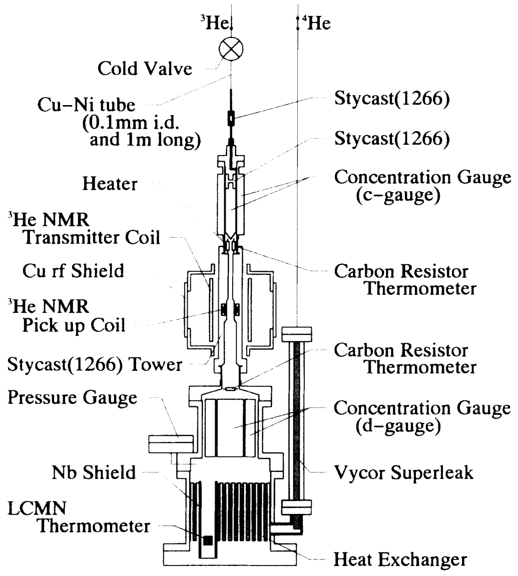

FIG. 1. Schematic drawing of the sample cell.

about 200 g of sintered silver powder. Its surface area is about 250 m^2. A capacitance-type concentration gauge of 0.5-mm gap is positioned so that it is always wholly immersed in the dilute phase; so we call it the d-gauge. The pressure gauge is a capacitance-type one. At the top of the cell we have another concentration gauge of 0.2-mm gap. Before starting the pressurization process it is filled with the concentrated phase; we call it the c-gauge. The ^{3}He line is connected to the top of the c-gauge from a cold valve [10] fastened to the mixing chamber. The total effective volume for the liquid is about 77 cm^3, almost half of which comes from the sintered-silver region. Because of the cold valve and the superleak, the total amount of ^{3}He in the sample cell is kept constant and the liquid in the cell has no direct connection to the liquid-vapor interface. The latter is a necessary condition to eliminate the possibility of the nucleation of the ^{3}He concentrated phase from the interface [11].

In addition to the three thermometers inside of the cell, we have a Pt-NMR thermometer fastened to the nuclear stage, which is used for the determination of the liquid temperature below about 5 mK because it has a much larger contact area with the liquid than the LCMN thermometer. The liquid temperature was related to the temperature with the Pt thermometer by measuring the nuclear free-induction signal of the demixed ^{3}He phase through and below the superfluid transition at various pressures, during both the cooling and the warming processes. The processes were performed so slowly that we obtained a good reproducibility in the relation. We used Greywall's scale [12].

First, the temperature dependences of the pressure and the concentration gauges are checked above 5.3 mK. Then pure ^{4}He is condensed into the cell to calibrate the gauges and thermometers. After that ^{3}He is condensed, during which ^{4}He flows out through the superleak. This procedure assures that the whole surface is completely covered with ^{4}He. After obtaining an appropriate amount of ^{3}He in the cell, we close the cold valve to keep it constant throughout the present experiment. Then, we stabilize the temperature and add ^{4}He continuously into the cell through the superleak to increase the pressure, during which the gauges and thermometers are monitored continuously. Every 2 min the data are recorded by a computer system. The flow rate of ^{4}He is about 4×10^{-6} mole/s, which corresponds to the pressure-change rate of about 0.25 kg/cm^{2}h. The increase of the amount of ^{4}He in the cell causes a transformation of the concentrated phase into the dilute phase due to both the increase of the ^{3}He solubility with pressure and the increase of the ratio of ^{4}He/^{3}He. So the phase-separation interface moves up. We notice that at P_u the interface reaches the top of the c-gauge [see Fig. 2(a)]. Then it gives the same ^{3}He concentration as the d-gauge.

The disappearance of the concentrated phase is recognized by both the d- and c-gauges as shown in Fig. 2(a), which clearly shows that the whole liquid enters into the unsaturated state at P_c. The solubility curve in the figure was calculated using available experimental data [5,13]. The nice fitting indicates the accuracy of our measurements. We stop the pressurization in the unsaturated region far from the solubility curve. Above about 20 mK we should wait for hours at this point until we obtain a homogeneous state in the whole liquid [7,8], which will be discussed in a separate paper.

After obtaining the equilibrium state in the unsaturated region, we start the depressurization process. The pressure-change rate is about 0.25 kg/cm^{2}h. It is almost 10^4–10^5 times slower than the decomposition experiments near the tricritical point [14]. In this process, as can be seen in Figs. 2(b) and 2(c), the homogeneity of the concentration is almost perfect in all temperature regions until we reach the demixing point P_d, which is noticed by the abrupt changes in the concentration gauges and in thermometers. Near the demixing point we mostly measured only the concentration and the pressure gauges in order to diminish unnecessary disturbances as much as possible. As seen in Figs. 2(b) and 2(c) the abrupt changes in both concentration gauges occur almost simultaneously. These gauges are separated about 120 mm in distance and the narrowest path is 3 mm in diameter and 40 mm long. Therefore, we may say that the demixing occurs almost simultaneously in the whole liquid. The jump of the ^{3}He concentration of the dilute phase at P_d is obtained from the data of the d-gauge and is denoted by $\Delta x_{3,\mathrm{cr}}$ which is the critical supersaturation we want to obtain. In Fig. 3 are plotted the thus obtained values of $\Delta x_{3,\mathrm{cr}}$ as a function of temperature.

From Fig. 3 we can see that the critical supersaturation becomes almost independent of temperature below about 10 mK, while it increases with temperature above about 10 mK. This temperature-independent behavior suggests

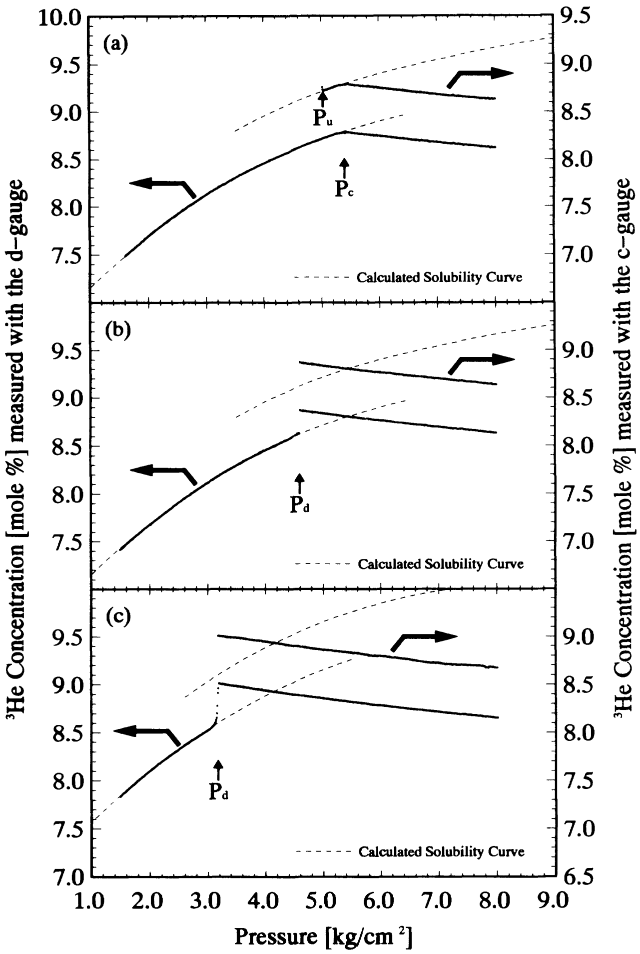

FIG. 2. Change of the ^{3}He concentration as a function of pressure: (a) pressurization process at 5.3 mK, (b) depressurization process at 5.3 mK, and (c) depressurization process at 80 mK.

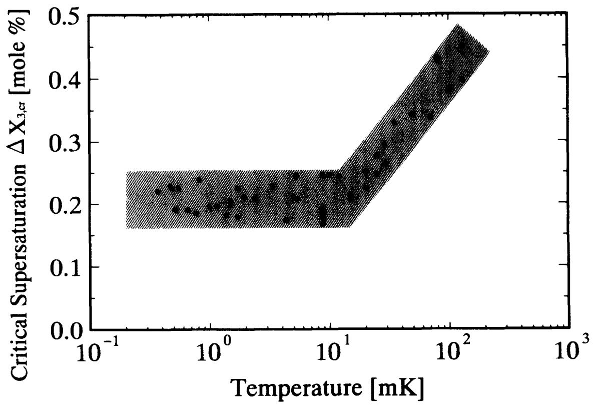

FIG. 3. The critical supersaturation of ^{3}He, $\Delta x_{3,\mathrm{cr}}$, as a function of temperature. The hatching is only to guide the eye.

that we are in the quantum regime below about 10 mK. Although this temperature is very close to the crossover temperature predicted by Lifshitz, Polesskii, and Khokhlov, the value of $\Delta x_{3,\mathrm{cr}}$ is more than 1 order of magnitude smaller than their estimate. There is some ambiguity in the estimation by Lifshitz, Polesskii, and Khokhlov for the chemical potential of ^{3}He in the metastable state. However, it seems difficult to attribute this big difference to that ambiguity. Since the crossover temperature is related to the amount of $\Delta x_{3,\mathrm{cr}}$ [3], we should say that the present experimental result is not consistent with the theory of Lifshitz, Polesskii, and Khokhlov. At present we have no clear explanation for this inconsistency. It may reinforce Leggett's viewpoint about the statistical mechanics of very improbable processes [15]. It is interesting to note that Landau *et al.* [6] reported $\Delta x_{3,\mathrm{cr}}$ of about 0.3% around 30 mK at 0.53 atm, which is comparable with our present data. Recently, Mikheev *et al.* [16] reported the result of supersaturation in the temperature region 50–200 mK. Their method is completely different from ours and their $\Delta x_{3,\mathrm{cr}}$ looks about a factor of 2 larger than ours in the overlapping temperature region, but a detailed comparison is not possible because they did not give their experimental data for the equilibrium solubility after demixing to obtain the values of $\Delta x_{3,\mathrm{cr}}$.

The increase of $\Delta x_{3,\mathrm{cr}}$ with temperature above about 10 mK conflicts with a simple expectation of the crossover consideration from the quantum to the classical regime. Recently, Burmistrov and Dubovskii [17] reconsidered the decomposition of a supersaturated ^{3}He-^{4}He mixture in the same way as Lifshitz, Polesskii, and Khokhlov but by taking the dissipation effect into account. They showed that the dissipations due to various kinetic processes suppress the decay of the supersaturated state. Although we cannot at present make a direct comparison between our results and their expression for the frictional coefficient, the ^{3}He mass diffusion seems dominant. If the ^{3}He atom diffusion is not enough, the droplet stops growing. The diffusion constant has a very steep temperature dependence, $1/T^5$ [18]. This might be one possibility to explain the curious temperature dependence of our $\Delta x_{3,\mathrm{cr}}$. Another possibility we are considering is the effect of the change of the state of ^{4}He in the nucleation process. The nuclei in the high-temperature region are not the pure ^{3}He phase as considered by Lifshitz, Polesskii, and Khokhlov but contain a small amount of ^{4}He, which increases with temperature. The ^{4}He in the supersaturated state is in the superfluid state while in the nuclei it is in the normal state. It is not obvious whether such a change of state proceeds smoothly. Comparing Figs. 2(b) and 2(c), we notice that the time necessary for the d-gauge to come back to the equilibrium solubility curve after demixing is much longer at 80 mK. Such a tendency becomes clear above about 40 mK, which may be related to the mechanisms that cause the increase of $\Delta x_{3,\mathrm{cr}}$ in the high-temperature region.

From Fig. 3 we see that the rapid nucleation points form a band of rather large width. It is not a sharp line as considered by Lifshitz, Polesskii, and Khokhlov. We believe that the scatter of the points is due to some intrinsic stochastic nature of the nucleation process, because good homogeneity is attained and the process is performed very slowly. It is noted, however, that in spite of all the precautions taken in the present study, one cannot completely exclude the possibility of an effect of heterogeneous nucleation on the observed results. It is desirable to make a quantitative estimation about the inertness of the wall covered with ^{4}He for the nucleation of the ^{3}He concentrated phase. It is an interesting future subject to study the lifetime of the metastable state at various stages of supersaturation near the rapid nucleation region determined in the present study.

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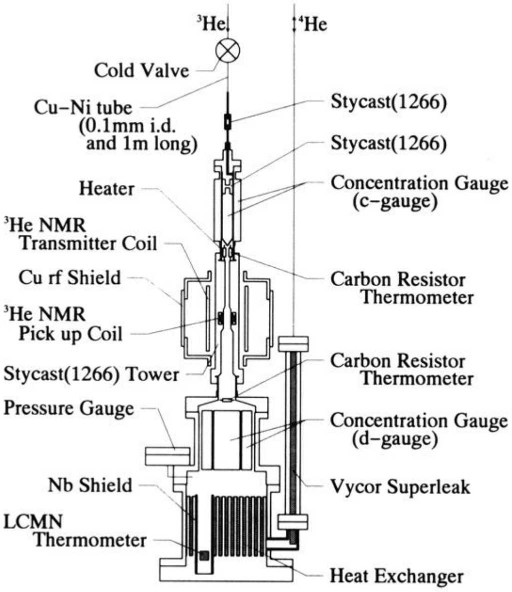

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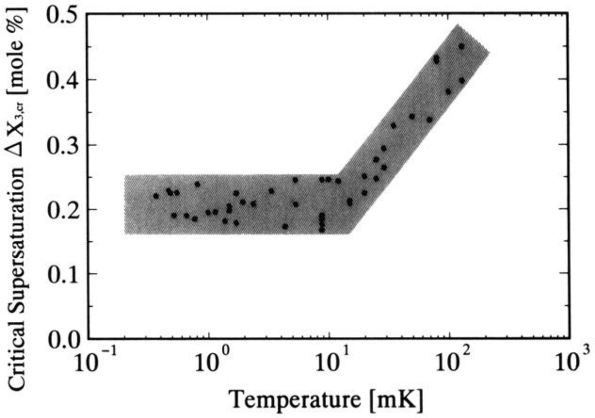

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