

On the Thermodynamic Equilibrium in the ^3He –Aerogel System at Low Temperatures

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A new method for studying the processes of the establishment of the thermodynamic equilibrium in the adsorbed ^3He layers in highly porous media has been proposed. Using this method, the thermalization of adsorbed ^3He on silica aerogel at a temperature of 1.5 K has been studied. The process of the establishment of the thermodynamic equilibrium has been controlled by measuring the pressure in an experimental cell, the amplitude of the NMR signal, and the nuclear spin–spin and spin–lattice relaxation times of adsorbed ^3He . It has been shown that the establishment of the thermodynamic equilibrium in the adsorbed ^3He –aerogel system is characterized by a time of 26 min.

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The study of the ^3He –aerogel system in recent 15 years has been of interest in view of the effect of disorder induced by the aerogel strands on the properties of superfluid ^3He phases [1–4]. However, the spin kinetics of ^3He in contact with aerogel above the Fermi degeneracy temperature (about 0.5 K) demonstrates a series of quite interesting features due to the considerable effect of the layer adsorbed on the aerogel surface on the nuclear magnetic relaxation of liquid ^3He [5, 6]. For the correct interpretation of the observed processes of nuclear magnetic relaxation at low and ultralow temperatures, it is of interest to study the processes of the establishment of the thermodynamic equilibrium in the ^3He –aerogel system. It is known that the thermal conductivity of aerogel at low temperatures is by orders of magnitude less than that of bulk SiO_2 ; this relation is probably the manifestation of the fractal structure of the aerogel [7]. The processes of the establishment of the thermodynamic equilibrium in liquid ^3He in the normal phase in contact with aerogels at ultralow temperatures were studied earlier in [8], where it was shown that the thermal conductivity of the system is determined by the thermal conductivity of liquid ^3He and that the effect of the aerogel structure starts to be manifested at ultralow temperatures. In addition, the model of the inhomogeneous potential of the adsorption of ^3He atoms on the surface of aerogel strands, which was proposed in [5] and supported by the recent quantum-chemical calculations [9], also leads to the necessity of studying such processes in detail. In principle, the temperature of the aerogel matrix could be determined from the intensi-

ties of the ^{29}Si and ^{17}O NMR nuclear magnetic resonance (NMR) signals. However, it is difficult to implement such a method, due to the low natural abundances of these isotopes (4.7 and 0.037%, respectively) and long spin–lattice relaxation times [10]. The paramagnetic centers in the aerogel strands induced by X-ray radiation can serve as good probes for studying the processes of the establishment of the thermodynamic equilibrium and determining the aerogel temperature at low temperatures [11], since the intensity of the EPR signal is, in most cases, inversely proportional to the absolute temperature of a sample. However, the concentration of the intrinsic paramagnetic centers in the nonirradiated aerogels is low and the EPR signal is usually not observed. In this work, we propose a method for studying the processes of the establishment of the thermodynamic equilibrium in the ^3He –aerogel system from the results of measuring the nuclear magnetic resonance parameters of ^3He on the surface of the nonirradiated SiO_2 silica aerogel with a filamentary structure in which the paramagnetic centers are absent. An aerogel sample (95%) of a cylindrical shape ($d = 5$ mm and $h = 12$ mm) was placed in a glass (Pyrex) ampoule, which was put in a helium (^4He) cryostat. The ampoule was sealed leak tightly to the ^3He gas handling system. The aerogel sample was evacuated (to 0.01 mbar) for 24 h at room temperature and was pre-cooled for 2 h at a temperature of 1.5 K in the ^4He cryostat directly before an experiment. A tank with ^3He ($V = 8$ cm³, $P = 6$ mbar, $T = 300$ K) was joined to the evacuated sample via a capillary (internal diameter $d = 3$ mm) about 3 m long.

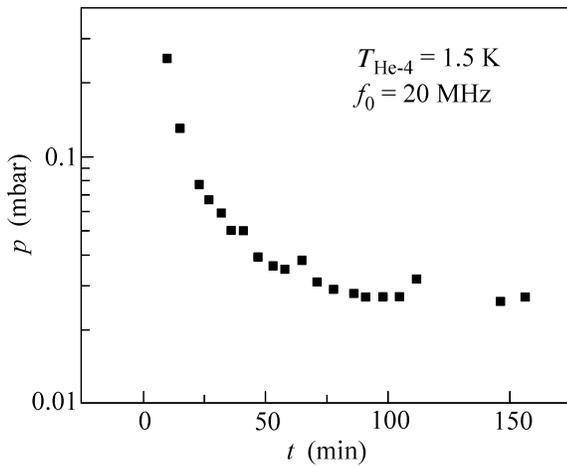


Fig. 1. Pressure of gaseous ^3He versus the time after the beginning of condensation.

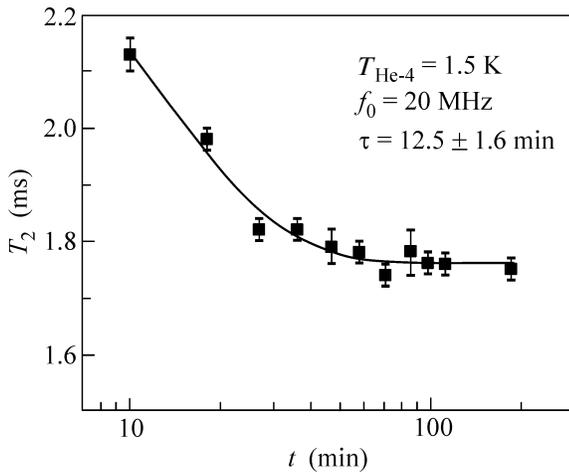


Fig. 2. Dependence of the ^3He relaxation time T_2 on the time after the beginning of condensation. The solid line is the fit by Eq. (1).

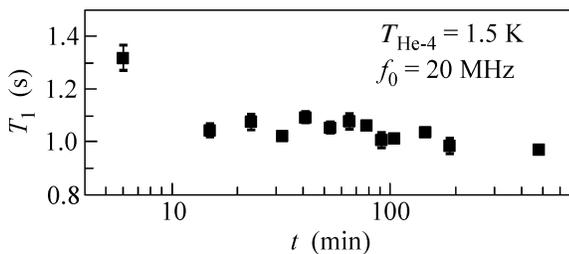


Fig. 3. Dependence of the ^3He relaxation time T_1 on the time after the beginning of condensation. The characteristic time τ in exponential fit (1) of the establishment of the equilibrium value of the relaxation time is less than 15 min.

It was assumed that the entire amount of ^3He passed to the NMR cell in 6 min and the pressure did not exceed 1 mbar. The amount of ^3He necessary to fill the adsorbed layer on the aerogel surface at the given temperature was chosen according to the method described in [5]. A homebuilt spectrometer with the frequency range from 3 to 50 MHz was used to measure the ^3He NMR signal amplitudes and nuclear magnetic relaxation parameters. The spin–lattice relaxation times were measured by the saturation recovery method using the amplitude of the free induction decay signal after the saturation pulse. The spin–spin relaxation times were measured by the Hahn method. The amplitude of the NMR signal was measured using the spin echo signal. All measurements were performed at a ^3He Larmor frequency of 20 MHz, the duration of the 90° pulse was $2.8 \mu\text{s}$, and the delay between the 90° and 180° pulses was $100 \mu\text{s}$. A temperature of 1.5 K was achieved by the evacuation of ^4He vapors from the cryostat. The time dependence of the pressure of ^3He during the thermalization process in the ^3He –aerogel system is shown in Fig. 1. A rough fit of these experimental data by exponential decay gives a characteristic time of about 15 min. It is assumed that this time is the characteristic time of the simultaneous adsorption of ^3He atoms on the aerogel surface and thermalization in the adsorbed ^3He layer. Since this pressure was measured in the “hot” part of the ^3He line, there was the thermomolecular pressure difference [12], and the pressure in the experimental cell differed from the measured value below 0.1 mbar. Therefore, the obtained characteristic time can be only the estimated one. The thermalization process in the ^3He –aerogel system is also manifested in the dependences of the ^3He nuclear spin–spin and spin–lattice relaxations on the time of the experiments (Figs. 2 and 3). If the relaxation times approach the equilibrium values (at $T = 1.5$ K) according to the exponential law

$$A = A_0 + A_1 e^{-t/\tau}, \quad (1)$$

then the performed approximations indicate that the τ parameter is about 12.3 min for the line in Fig. 2 and is less than 15 min for the dependence in Fig. 3. However, it is impossible to reliably establish the functional dependence in Fig. 3 because of the small number of the experimental points in the curve in the range from 6 to 15 min due to the long time of measuring the T_1 parameter and the low signal-to-noise ratio. It should be noted that the functional dependences of T_1 and T_2 times on the time after the beginning of ^3He condensation in aerogel are determined not only by the processes of the redistribution of the ^3He molecules in the adsorbed layer, but also by the temperature dependence of these parameters (under the equilibrium conditions). The intensity of the NMR signal can also serve as an indication of the thermalization of ^3He in

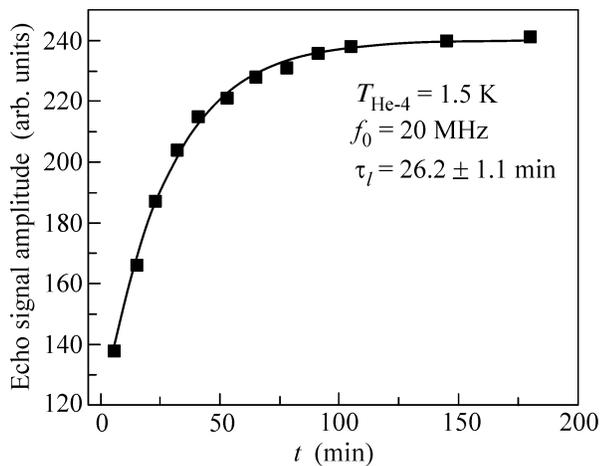


Fig. 4. Intensity of the ^3He NMR signal in aerogel on the time after the beginning of condensation. The solid line is the fit by Eq. (1).

aerogel, since the intensity of the NMR signal of adsorbed, liquid, and gaseous ^3He above the Fermi degeneracy temperature is inversely proportional to the temperature (under the equilibrium conditions). The dependence of the intensity of the ^3He NMR signal on the time after the beginning of ^3He condensation into the cell is shown in Fig. 4. The curve in Fig. 4 can be approximated by Eq. (1) with the τ value of about 26 min. Since this characteristic time is the longest obtained value, it most adequately characterizes the thermalization process in this system. It should be noted that all experimental data given in Figs. 1–4 describe the global thermalization process in the ^3He –aerogel system and they are the manifestations of different physical processes occurring in the system (adsorption, redistribution of atoms in the adsorbed layer, thermalization of the aerogel layer and matrix, and general equalization of the system temperature with the ^4He bath temperature). To study these contributions more impartially and in more detail, it is necessary to considerably modify the experimental setup and experimental method.

Thus, it follows from the given experimental data that the thermalization process in the ^3He –adsorbed aerogel system with the ^4He bath is characterized by a time of 26 min. In this work, a particular case of the ^3He –aerogel system was considered, namely, the pro-

cesses of the establishment of the thermal equilibrium of the ^3He layer adsorbed on the surface. In the case of the appearance of free ^3He atoms (of the liquid or gas phase), these processes are considerably accelerated due to the fast mass diffusion and energy exchange with the NMR cell walls. For example, when the ^3He pressure over the surface of the adsorbed layer is higher than 5 mbar, the thermalization time was less than 5 min. As a result, the NMR method of studying the processes of the establishment of the thermal equilibrium in the adsorbed ^3He layers in highly porous media was proposed. This method was used to study the thermalization of ^3He adsorbed in silica aerogel at $T = 1.5$ K.

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