The spin kinetics of ^{3}He in contact with nanosized crystalline powders LaF$_3$

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Abstract. The spin kinetics of ^{3}He in contact with nanosized crystalline powders LaF$_3$ has been studied by NMR methods at the temperature 1.5 K. The ^{3}He longitudinal relaxation time increases proportionally to the magnitude of the external magnetic field and the transverse relaxation time does not depend on the magnetic field. Relaxation of the gaseous and liquid ^{3}He in contact with nanosized crystalline powder LaF$_3$ takes place by the ^{3}He adsorbed layer. The nuclear magnetic relaxation of adsorbed ^{3}He layer on the surface of LaF$_3$ nanoparticles is due to the two-dimensional spin-diffusion motion.

Introduction

Hyperpolarized ^{3}He gas is widely used in neutron accelerators for the polarization of neutron beams [1–4], nuclear magnetic resonance tomography, and diverse fundamental scientific researches [5–10]. Therefore, obtaining the hyperpolarized spin state of ^{3}He nuclei using new methods remains a topical problem to date.

In 1995 the possibility of dynamic ^{3}He nuclear polarization using dielectric Van Vleck paramagnetic was discussed [11]. The channel of energy transfer from the PrF$_3$ to liquid ^{3}He via magnetic coupling between the nuclear spins was proposed. The "^{141}Pr - ^{3}He" system has been studied in detail [12-16].

The process previously interpreted [13] as a relaxation mechanism of ^{3}He on the surface of nanosized crystalline powders PrF$_3$ through the adsorbed layer may be a cross-relaxation effect between ^{141}Pr and ^{3}He nuclei. To confirm this idea the additional experiments with nanosized crystalline powders LaF$_3$ (diamagnetic analogue of PrF$_3$) have been required.

We report here the experimental investigation of ^{3}He spin kinetics in the system "LaF$_3$ nanoparticles - ^{3}He". The home built NMR spectrometer has been used and low temperature was achieved by ^{4}He vapor pumping. The complete details of the experimental setup were published earlier [17]. The typical thermalization time of ^{3}He adsorbed layer on a diamagnetic solid state substrate is less than 2 hours for our setup and temperature range [18]. The covering of sample surface by ^{3}He was carried out in the following sequence: gaseous ^{3}He was condensed in the experimental cell at temperature 1.5 K in small amounts (about 0.5 cm^3 STP). The pressure in the system has been monitored, and when it reached a value less than 10^{-2} mbar the next dose of ^{3}He was added. When the equilibrium pressure in the system rose above 10^{-1} mbar, the entire surface was assumed to be covered by ^{3}He layer. Further condensation of ^{3}He led to an increase of the pressure with most of the ^{3}He remaining in the gas phase. When the pressure approached the vapor saturation value (67 mbar at

1.5 K), the liquid phase appeared in the cell and further dosing of ^{3}He was controlled by the ^{3}He NMR signal amplitude until whole sample has been filled completely. Before all NMR measurements the sample was thermalized during 2 hours.

Results and discussion

The series of LaF$_3$ nanosamples were synthesized using different microwave irradiation time (sample 1 – 0 min, sample 2 – 20 min, sample 3 – 40 min, sample 4 – 60 min, sample 5 – 420 min), the procedure described in [13]. The X-ray analysis of synthesized samples is shown in figure 1, which identifies samples as crystalline powders LaF$_3$. Further on, the data on ^{3}He spin kinetics in the system "LaF$_3$ nanoparticles - ^{3}He" are presented for sample 2.
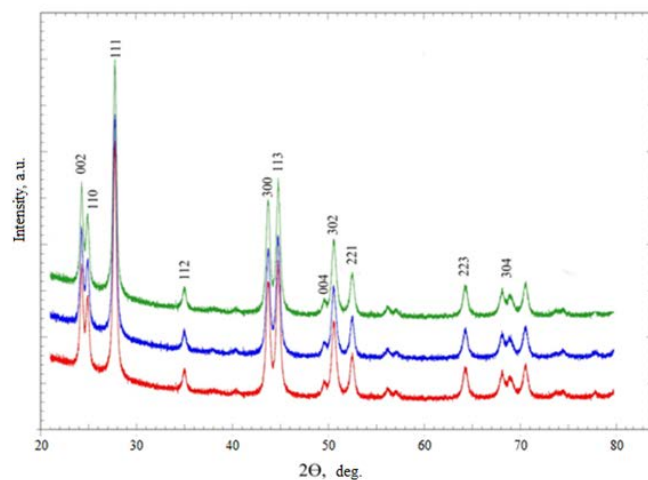

Figure 1. X-ray analysis of LaF$_3$ nanoparticles.

The typical image of PrF$_3$ (diamagnetic analog of LaF$_3$) nanoparticle obtained by high resolution transmission electron microscopy presented in figure 2. The size of nanoparticles is 20-40 nm.
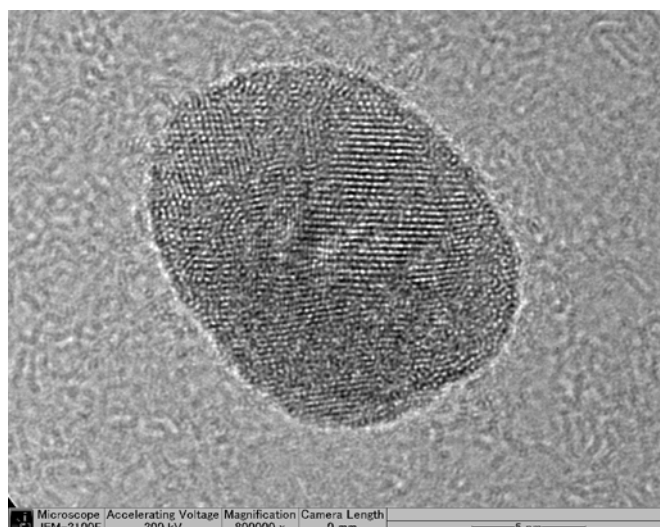

Figure 2. High-resolution transmission electron microscopy image of PrF$_3$ nanoparticle.

The magnetic field dependence of the ^{3}He nuclear magnetic relaxation times in contact with LaF$_3$ nanoparticles in different aggregate states of ^{3}He have been investigated. Dependences of the ^{3}He relaxation times T_1 and T_2 on the magnetic field for the three aggregate states are shown in figures 3 and 4.
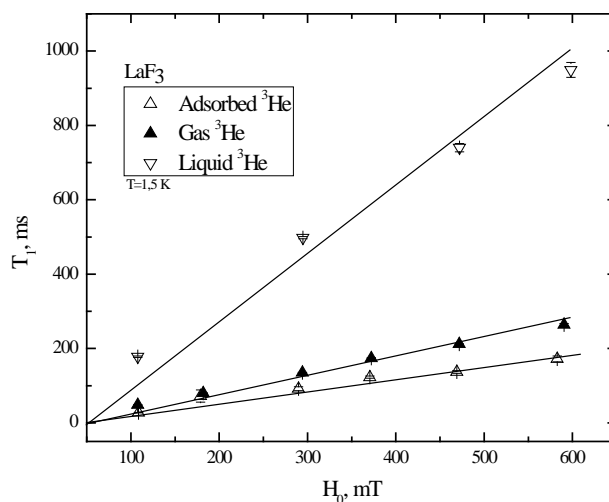

Figure 3. Longitudinal relaxation time of ^{3}He in contact with LaF$_3$ nanoparticles in different aggregate states of ^{3}He versus magnetic field.

It is seen in figure 3 that the longitudinal relaxation time T_1 increases proportionally to the magnitude of the external magnetic field. The transverse relaxation time T_2 is independent on the applied magnetic field (figure 4).
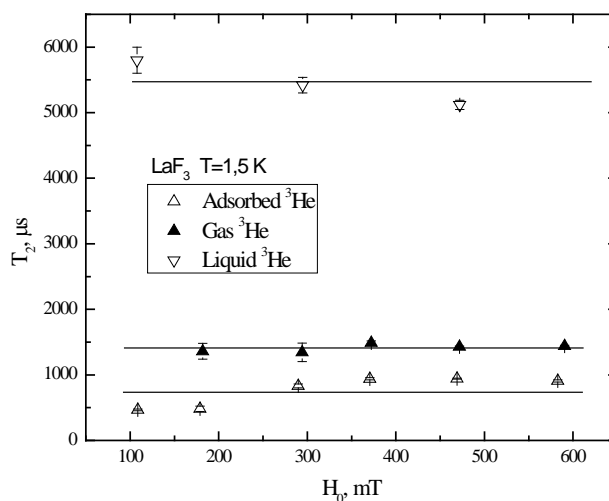

Figure 4. Transverse relaxation time of ^{3}He in contact with LaF$_3$ nanoparticles in different aggregate states of ^{3}He versus magnetic field.

The presence of "free" ^{3}He (gaseous or liquid) above adsorbed layer in the experimental cell significantly increase the relaxation times values, therefore the relaxation occurs through the ^{3}He adsorbed layer [19].

The ^{3}He T_1 and T_2 dependences in the gaseous and liquid phases in the cell on the total number of ^{3}He atoms (figures 3 and 4) clearly shows that the magnetic relaxation times are directly proportional to the corresponding relaxation times in the adsorbed layer and to the ratio of the number of ^{3}He spins of whole nuclear spin system to the number of ^{3}He spins in the layer, i.e.

$$T_1 = T_{1S} \cdot N_0/N_S, \tag{1}$$

where T_1, T_{1S} – longitudinal magnetization recovery times, N_0, N_S-number of spins in the whole system and the adsorbed layer, respectively;

$$T_2 = T_{2S} \cdot N_0/N_S, \tag{2}$$

where T_2, T_{2S} - times of the transverse magnetization decay, N_0, N_S-number of spins in the entire system and in the adsorbed layer, respectively.

This fact as well as the character of dependence of the relaxation times on the magnetic field plays in favor of relaxation mechanism of adsorbed ^{3}He in the two-dimensional film by the two-dimensional spin-diffusion motion, as proposed by Cowan [20, 21].

The model of 2D relaxation, proposed by Cowan, only qualitatively describes obtained data. Comparison of ^{3}He T_1 values of adsorbed layer on various diamagnetic surfaces [21-24] shows quite good similarity with presented here experimental data, which additionally supports proposed relaxation mechanism. However, there are several additional mechanisms of ^{3}He surface relaxation due to dipole-dipole interaction between ^{3}He spins and solid substrate [25-31]. The direct transfer of magnetization from ^{3}He to ^{19}F will not speed up ^{3}He nuclear transverse relaxation sufficiently to observed values, because at helium temperatures the intrinsic spin-lattice relaxation of ^{19}F in pure diamagnetic fluoride crystals is in the order of 100s of seconds [see for example 32]. The ^{3}He nuclear magnetic relaxation of adsorbed layer due to the modulation of the dipole-dipole interaction between ^{19}F and ^{3}He by the quantum zero point motion [31] is not sufficient, as well.

Conclusion

Nuclear magnetic relaxation of the gaseous and liquid ^{3}He in contact with nanosized crystalline powder LaF$_3$ takes place by the adsorbed layer. The nuclear magnetic relaxation of adsorbed ^{3}He layer on the surface of LaF$_3$ nanoparticles is due to the two-dimensional spin-diffusion motion.

Acknowledgments

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