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The spin kinetics of ³He in contact with nanosized crystalline powders LaF₃

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Abstract. The spin kinetics of ³He in contact with nanosized crystalline powders LaF₃ has been studied by NMR methods at the temperature 1.5 K. The ³He 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 ³He in contact with nanosized crystalline powder LaF₃ takes place by the ³He adsorbed layer. The nuclear magnetic relaxation of adsorbed ³He layer on the surface of LaF₃ nanoparticles is due to the two-dimensional spin-diffusion motion.

Introduction

Hyperpolarized ³He 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 ³He nuclei using new methods remains a topical problem to date.

In 1995 the possibility of dynamic ³He nuclear polarization using dielectric Van Vleck paramagnetic was discussed [11]. The channel of energy transfer from the PrF₃ to liquid ³He via magnetic coupling between the nuclear spins was proposed. The "¹⁴¹Pr - ³He" system has been studied in detail [12-16].

The process previously interpreted [13] as a relaxation mechanism of ³He on the surface of nanosized crystalline powders PrF₃ through the adsorbed layer may be a cross-relaxation effect between ¹⁴¹Pr and ³He nuclei. To confirm this idea the additional experiments with nanosized crystalline powders LaF₃ (diamagnetic analogue of PrF₃) have been required.

We report here the experimental investigation of ³He spin kinetics in the system "LaF₃ nanoparticles - ³He". The home built NMR spectrometer has been used and low temperature was achieved by ⁴He vapor pumping. The complete details of the experimental setup were published earlier [17]. The typical thermalization time of ³He 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 ³He was carried out in the following sequence: gaseous ³He was condensed in the experimental cell at temperature 1.5 K in small amounts (about 0.5 cm³ STP). The pressure in the system has been monitored, and when it reached a value less than 10⁻² mbar the next dose of ³He was added. When the equilibrium pressure in the system rose above 10⁻¹ mbar, the entire surface was assumed to be covered by ³He layer. Further condensation of ³He led to an increase of the pressure with most of the ³He remaining in the gas phase. When the pressure approached the vapor saturation value (67 mbar at

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1.5 K), the liquid phase appeared in the cell and further dosing of ³He was controlled by the ³He 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 LaF₃ 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 LaF₃. Further on, the data on 3 He spin kinetics in the system "LaF₃ nanoparticles - 3 He" are presented for sample 2.

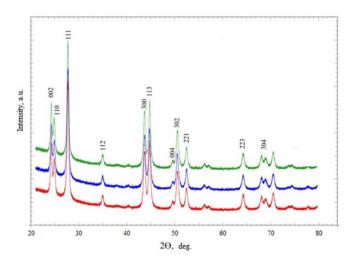


Figure 1. X-ray analysis of LaF₃ nanoparticles.

The typical image of PrF₃ (diamagnetic analog of LaF₃) nanoparticle obtained by high resolution transmission electron microscopy presented in figure 2. The size of nanoparticles is 20-40 nm.

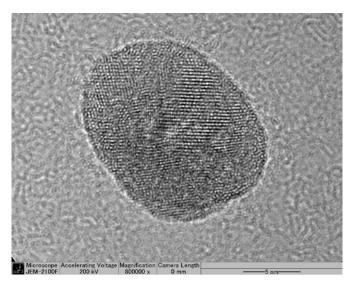


Figure 2. High-resolution transmission electron microscopy image of PrF₃ nanoparticle.

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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.

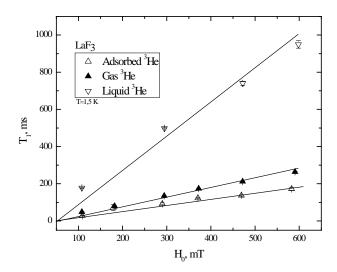


Figure 3. Longitudinal relaxation time of ³He in contact with LaF₃ nanoparticles in different aggregate states of ³He versus magnetic field.

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).

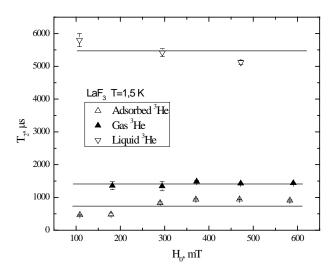


Figure 4. Transverse relaxation time of ³He in contact with LaF₃ nanoparticles in different aggregate states of ³He versus magnetic field

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

The ³He T₁ and T₂ dependences in the gaseous and liquid phases in the cell on the total number of ³He 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 ³He spins of whole nuclear spin system to the number of ³He 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 ³He 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 ³He T₁ 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 ³He surface relaxation due to dipole-dipole interaction between ³He spins and solid substrate [25-31]. The direct transfer of magnetization from ³He to ¹⁹F will not speed up ³He nuclear transverse relaxation sufficiently to observed values, because at helium temperatures the intrinsic spin-lattice relaxation of ¹⁹F in pure diamagnetic fluoride crystals is in the order of 100s of seconds [see for example 32]. The ³He nuclear magnetic relaxation of adsorbed layer due to the modulation of the dipole-dipole interaction between ¹⁹F and ³He 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 LaF₃ takes place by the adsorbed layer. The nuclear magnetic relaxation of adsorbed 3 He layer on the surface of LaF₃ nanoparticles is due to the two-dimensional spin-diffusion motion.

Acknowledgments

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