The Study of the System “Van Vleck Paramagnet PrF₃ - Helium-3”

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Abstract. Present paper is the result of experimental investigations of the system “Van Vleck paramagnet PrF₃ – liquid ³He”, where earlier cross-relaxation effect between liquid ³He nuclei and \(^{141}\)Pr nuclei was discovered. We report on \(^{141}\)Pr spin kinetics in Van Vleck paramagnet crystal powders PrF₃ with different paramagnet impurities. All experiments have been made by pulse NMR methods at temperature 1.5 K.

1. Introduction

Magnetic dipole interaction between the nuclear spins of liquid ³He and the nuclear spins of a solid substrate between ³He and \(^{19}\)F nuclei in adsorbed ³He-polytetrafluoroethylene (DLX-6000) system was discovered in 1981 [1]. Dominant relaxation channel of the \(^{19}\)F nuclei is their dipole interaction with the ³He nuclei [1,2]. Afterwards, the effect of cross-relaxation with the nuclei of liquid ³He was observed in several other systems [3-6]. Cross-relaxation effect between liquid ³He nuclei and \(^{141}\)Pr of PrF₃ crystal powder by authors was observed first time [7].

In our experiments, investigations of \(^{141}\)Pr and ³He nuclei magnetic properties were made by pulse NMR method at temperature 1.5 K and frequency 6.63 MHz. In experiments, the crystal powder PrF₃ with medium size 10-45 mkm was used as a sample. Field dependences of \(^{141}\)Pr nuclei relaxation times were studied in systems: “PrF₃ powder - liquid ³He” and “PrF₃ powder - liquid ³He”.

Effect of the resonance magnetic coupling is the following. Obtained field dependence of \(T₁\) of \(^{141}\)Pr nuclei is bell-shaped, with the centre in magnetic field that corresponds to the peak value of ³He nuclei NMR signal when longitudinal magnetic relaxation time of \(^{141}\)Pr nuclei becomes twice longer than on the wings. In this work we continue previous investigations of magnetic relaxation properties of nuclei in those systems with another PrF₃ crystal powder. Especially, we need to know information about mechanisms of \(^{141}\)Pr magnetization relaxation in PrF₃. Study of this process is additionally complicated because there is no theory for magnetisation relaxation of Van Vleck nuclei with nuclear spin \(I > 1/2\) [8].
2. Samples and Experiment
The PrF$_3$ crystal powder with medium size 10-45 mkm was used as a sample N1. The EPR data shows that the total quantity of paramagnetic admixture (Nd$^{3+}$, Gd$^{3+}$, Er$^{3+}$, Dy$^{3+}$) in PrF$_3$ was less than 0.02\% (see fig. 1). As a sample N2, PrF$_3$ crystal powder with same size and paramagnetic admixture less than 0.01\% was used (see fig. 2).

![EPR spectra of sample N1 at 77 K](image1)

![EPR spectra of sample N2 at 50 K](image2)

Fig. 1. EPR spectra of sample N1 at 77 K  
Fig. 2. EPR spectra of sample N2 at 50 K

According to mass-spectroscopy measurements mole concentration of $^4$He in $^3$He was less than 0.05\%. Our investigations of $^{141}$Pr nuclei magnetic properties were made by pulse NMR method at temperature 1.5 K and frequency 6.65 MHz. The longitudinal magnetic relaxation time for the nuclei has been measured using the $\pi/2 - t - \pi/2 - t' - \pi$ pulse sequence (t is the delay between the saturating $\pi/2$ pulse and the readout pair, t’ is a constant during measurements).

![Relaxation of the $^{141}$Pr nuclei in the "PrF$_3$ powder–liquid $^3$He" system](image3)

Fig. 3. Relaxation of the $^{141}$Pr nuclei in the "PrF$_3$ powder–liquid $^3$He" system (filled circles) and "PrF$_3$ powder–liquid $^4$He" (opened circles) at 1.5 K at a frequency of 6.65 MHz for sample N1.
3. Results

As was mentioned before, in this system cross-relaxation was discovered first time (fig 3). The effect was in transfer of magnetization from $^3$He spins trough the surface to $^{141}$Pr spins. Spin lattice relaxation time for $^{141}$Pr has been increased in 2 times at cross relaxation conditions.

The spin-lattice relaxation of $^{141}$Pr is the matter of interest by itself, because there is no theory, explaining experimental results [8]. Further investigation on a new sample N2 shows different behaviour and mainly the spin kinetic of $^{141}$Pr changed. Longitudinal magnetization relaxation of $^{141}$Pr nuclei was measured in both samples in a magnetic field 195 mT (fig. 4).

For sample N1, relaxation easily approximates with function:

$$A(t) = A(\infty) \left(1 - B \cdot e^{-(t/T_1)^{0.5}}\right)$$

(1),

where parameters $T_1$ is longitudinal magnetic relaxation time of $^{141}$Pr nuclei, B determines saturation degree of $^{141}$Pr spin system due to the $\pi/2$ pulse, $A(t)$ is the amplitude of spin echo signal. Power 0.5 in an exponent corresponds to uniform $^{141}$Pr nuclei relaxation rates distribution in a powder sample [9].

The case of the sample N2 is more complicated. The longitudinal nuclear magnetization recovery is divided into 2 processes:

$$A(t) = A(\infty) \left(1 - A_1 \cdot e^{-(t/T_{11})^{0.5}} - A_2 \cdot e^{-t/T_{12}}\right)$$

(2),

where parameters $A_1$ and $A_2$ determines ratio between two relaxation processes, $T_{11}$ and $T_{12}$ are longitudinal magnetic relaxation times, $A(t)$ is the amplitude of spin echo signal of $^{141}$Pr nuclei.

![Fig. 4. Longitudinal magnetization relaxation of $^{141}$Pr nuclei in PrF$_3$ (sample N1 and sample N2) at 1.5 K at frequency of 6.65 MHz.](image)

Observed relaxation time of $^{141}$Pr nuclei in sample 1 is $T_1=7$ ms. In sample N2 with less paramagnetic impurities the same relaxation process is slower: $T_{11}=43.2\pm4.4$ ms and appears very slow relaxation component with $T_{12}=8.2\pm2.5$ s.

Very slow relaxation component $T_{12}$ makes experiments very complicated for cross-relaxation effect detection in comparison with sample N1. Simultaneously, during $T_1$ measurements, we observe 3 types of contribution into the signal: 2 relaxation components of $^{141}$Pr nuclei and 1 relaxation...
component of $^3$He nuclei. In order to split them correctly we need several hours measurement and experiment conditions are unstable during such a long time. For this reasons, observation of the cross-relaxation effect between $^{141}$Pr and $^3$He nuclei in system “PrF$_3$ – liquid $^3$He” is difficult in case of sample N2.

In order to continue our research, knowledge of connection between paramagnet impurities and $^{141}$Pr is very important. For describing paramagnet impurities effect on $^{141}$Pr nuclei relaxation we suggest to carry out measurements on PrF$_3$ single crystal.

4. Conclusions

Relaxation process of $^{141}$Pr nuclei magnetization in PrF$_3$ qualitatively depends on the amount of paramagnetic impurities. In this work we used powders as samples, because large surface area was necessary for successful observation of cross-relaxation effect. For accurate determining the effect of paramagnetic centers on Van Vleck nuclei magnetization relaxation and paramagnetic impurity concentration we suggest to use single crystals PrF$_3$ for this purpose.

Systematic pulse NMR investigations of $^{141}$Pr nuclei in single crystal PrF$_3$ at various temperatures, fields and orientations are required for determining the mechanism of $^{141}$Pr nuclei magnetic relaxation.

This work is the first step in experimental and theoretical research of NMR in Van Vleck paramagnets with nuclear spin I > 1/2.

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