EINSTEIN-DE HAAS NUCLEAR ANALOGUE EXPERIMENT[*]

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The large Compass polarized target at CERN was investigated as a nuclear model of the Einstein-de Haas magneto-mechanical experiment [1]. In this study the ^{3}He/^{4}He-dilution refrigerator cooled a sample of polarized irradiated ammonia (NH_3) down to 60 mK [2]. Because ^{3}He has a nuclear spin, ^{3}He-atoms in a magnetic field can transmit the angular momentum [3] from a polarized sample to the thermometers and the walls of the dilution chamber. At 0.03 T field and negative polarization, the cross-relaxation between proton ($I = 1/2$) and nitrogen ($I = 1$) spins produced the lattice vibrations with dominant relaxation of electrical nature. We observed and present here the nuclear magneto-mechanical effect and the spectra recorded by RuO-thermometers. This effect may explain the reduced relaxation time at the negative polarizations as compared with positive one in materials incorporating quadrupolar nuclei.

1. Introduction

The paper "Experimenteller Nachweis der Ampèreshen Molekularströne" by Einstein and W.J. de Haas [E-de H] was published in 1915 [1]. The concept of "Nuclear Spin" was elaborated later on in 1925 ÷1927. The main idea of [E-de H] experiment was that the natural ferromagnetism can be explained by the magnetic momentum of a closed Ampère current loop. To avoid a conflict between dissipative Ampère current and permanent natural magnetism, [E-de H] suggested that due to the absence of radiation in the natural magnets and according to the conservation of angular momentum law in the process of magnetizing and demagnetizing of a sample "*this change of the inner angular momentum must correspond to the occurrence of compensating angular momentum of an* ***another kind****; the latter will be a crude mechanical angular momentum*". 25 years after [E-de H] experiment, Gorter and Kahn [3] wrote "*The gyromagnetic effects … in a very direct way demonstrated the difference between electric dipoles, which consist of two equal charges of opposite signs at a short distance and magnetic dipoles, which consists of circular Ampère molecular currents*". This hint gave us the idea for nuclear analogue experiment, namely: the nuclear demagnetization, at certain conditions, must induce compensating mechanical angular momentum in the lattice. Almost hundred years later [E-de H] experiment we try a new way to study the nuclear magneto-mechanical effects, using superlow temperatures, Dynamic Nuclear Polarization (DNP) and the world largest polarized target.

2. Main Idea and Numeric Estimations

2.1. *Main idea of the experiment*

In solid dielectrics the energy of nuclear spins relaxes mainly through their interaction with paramagnetic impurities because the number of lattice phonons with nuclear frequencies is extremely small at low temperatures. Relaxation mechanism consists in modulating of the local magnetic field by means of thermal lattice vibrations and an excitation of nuclear transitions with the Fourier components of modulated field. In irradiated ammonia (NH_3), investigated below, the relaxation times come to thousands hours at 2.5 T and of the order of minutes at zero field at about 60 mK in both cases. Due to decreasing thermal capacitance of the lattice at lowering temperatures, one could expect

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that the energy of nuclear spins should also relax through mechanical vibrations of the lattice, analogously to the sample oscillations in the [E-de H] experiment [1]. Obviously, to identify the nuclear magneto-mechanical effect, one needs faster relaxation process than the process caused by electron impurities.

R. Pound showed [4] that relaxation rate of quadrupolar nuclei interacting with an electric field gradient enables to largely exceed the relaxation rate through impurities. If it is so at superlow temperatures, then the coupling of quadruple nitrogen nuclei with electric field gradients could excite of mechanical vibrations in the lattice.

2.2. *Numeric estimations*

Since magnetic momentum of proton of about one order larger than nitrogen one, we use the nuclear cross-relaxation in nonzero magnetic field to release the large energy of proton spins in NH_3 not through electron impurities but through the quadruple interactions of nitrogen spins with the lattice. In this case, the energy of magnetic dipolar interactions ($E_{d\text{-}d}$) between proton (μ_P) and nitrogen (μ_N) moments in (c.g.s) equals to

$$E_{d-d}=\frac{\mu_P\,\mu_N}{\left\langle r_{PN}^3\right\rangle_{d-d}}\approx\frac{1.4\cdot10^{-23}\cdot2.0\cdot10^{-24}}{\left\langle r_{PN}^3\right\rangle_{d-d}}\approx\frac{3.0\cdot10^{-47}}{\left\langle r_{PN}^3\right\rangle_{d-d}},\qquad(1)$$

where $\langle r_{PN}\rangle$ is the average distance between protons and nitrogen spins. Average quadrupolar energy of nitrogen spin (W_q) in the electric field gradient is [5]

$$\begin{aligned}W_q&=\frac{e^2Q}{4\cdot\left\langle r_N^3\right\rangle_q}\left\langle\left(3\cos^2\theta-1\right)^2\right\rangle_\theta\\&\approx\frac{\left(4.8\cdot10^{-10}\right)^2\cdot2.0\cdot10^{-26}}{4\cdot\left\langle r_N^3\right\rangle_q}\frac{4}{5}\approx\frac{9.0\cdot10^{-46}}{\left\langle r_N^3\right\rangle_q},\end{aligned}\qquad(2)$$

where eQ is the nitrogen quadruple moment, $\langle r_N\rangle_q$ is the average distance between charges of nucleus and electrons (e); θ is the angle between the principal axis of the field gradient tensor and the direction of magnetic field. Assuming that in the same material

$$\left\langle r_{PN}^3\right\rangle_{d-d}\approx\left\langle r_N^3\right\rangle_q$$

and using the classical theory of the relaxation [6] we obtain the ratio of quadrupolar to dipolar $T_q/\ T_{d\text{-}d}$ relaxation times as

$$\frac{T_q}{T_{d-d}}\approx\left(\frac{E_{d-d}}{W_q}\right)^2\approx\left(\frac{3.0\cdot10^{-47}}{9.0\cdot10^{-46}}\right)^2\approx0.001,\qquad(3)$$

If, for example, the nuclear relaxation time through the electron impurities is of about 1 hour, as it is in irradiated ammonia at about of 0.03 T and at 60 mK, then the quadrupolar relaxation time has of the order of seconds. We use this feature of quadruple relaxation to extract the nuclear magneto-mechanical effect from processes which involve the electron spins. Moreover, the conservation law of angular momentum demands that any conversion of the nuclear momentum must change not only spin energy $\langle Hq\rangle$ as well as the alignment $A(N)$ of nitrogen spin system. In fact, at zero magnetic fields the energy of nitrogen spin system is proportional to its alignment

$$\begin{aligned}\left\langle H_Q\right\rangle&=h\nu_Q\left\{3\cos^2\theta-1\right\}\left\langle3I_z^2-I\left(I+1\right)\right\rangle=\\&h\nu_Q\left\{3\cos^2\theta-1\right\}A\left(N\right)\end{aligned}\qquad(4)$$

where h is the Plank's constant, $\nu_Q=1/8\ (e^2qQ/h)$, (eq) is the value of the electric field gradient along the principal axis of the field gradient tensor, I and $\langle I_z\rangle$ are the spin and z-component of the angular momentum, correspondingly. It is obvious (see Introduction), that "*...angular momentum of an* ***another kind*** ..." or, to say, the transport of nuclear moments to the lattice mechanical vibrations should come due to varying $\langle I_z\rangle$ in $A(N\ )$. Both conditions are satisfied in our tests described below.

2.3. *Tree-cells Compass polarized target*

Our data were obtained with the Compass polarized target at CERN. It uses a powerful dilution refrigerator, a solenoid magnet with a longitudinal field of 2.5 T and a dipole magnet with a transverse field of 0.6 T. The target consists of three cells (30 + 60 + 30) cm long and 4 cm in diameter, filled with irradiated and granulated ammonia [2]. It operates with a microwave (MW) cavity (see Figure 1) in l = 4 mm wavelength range and in the temperature range from 0.06 to 0.25 K. MW-cavity consists of three cupper cells electrically isolated from each other by MW-stoppers.

The nuclear polarization is measured by ten commercial "Liverpool" Q-meters [7] connected to probing coils equally distributed along the target. The receiver circuits were permanently tuned to $\nu_0 = 106.42$ MHz and fed by a RF-synthesizer. It frequency was scanned by 1000 steps within 600 kHz bandwidth.

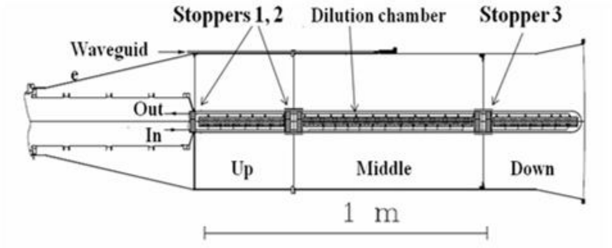

Figure 1. Three (Up, Middle and Down) microwave cells, powered through the waveguides. Electrical isolation between cells is performed using “Stoppers “ that provide free passage for He-flow. ^{3}He circulate through material from In to Outlet direction.

3. Experimental Results

3.1. *General results*

Detailed investigation of cross-relaxation in the ammonia at positive polarizations was done in [2] starting with high proton polarization of +89 % and nitrogen polarization of +16 %. The magnetic field was reduced to 0.045 T and raised back to 2.5 T several times. As a result, the nitrogen polarization was increased up to +40 %. It was also done the detail study of the line shape of nitrogen and proton spectra but the relaxation processes weren’t considered. In our study, at 2.5 T homogeneous solenoid field, the proton spins in ammonia were polarized by the DNP-method to ±80 %. Then tests were performed without use of external alternative MW and RF-fields. With the positive polarization, sweeping up and down of the static magnetic field up to 0.03 T does not affect thermometers. This means that the cross-relaxation at positive polarizations goes adiabatically without any visible lattice effects and the cross-relaxation produces only a partial exchange between spin species.

Nuclear magneto-mechanical effect is clearly observed at negative nuclear polarizations. Figure 2 shows thermal spectra recorded during the double passages of the cross-relaxation region nearby 0.03 T. These spectra come from the magneto-mechanical vibrations transporting from the lattice to liquid helium. This conclusion follows from the fact that the cells are electrically isolated from each other, and the design of our dilution chamber provides the free passage of helium flow in the dilution chamber. Such an explanation also confirms and almost identical line shape of spectra in different cells in Figure 2; it is clear, that vibrations are generated in the Middle cell with the highest started polarization and then the excitations are spread in the near-by cells. Figure 3 shows the spectra

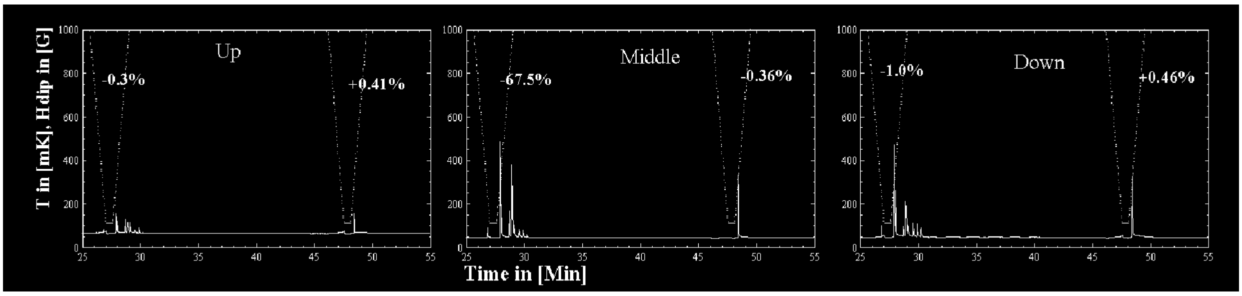

Figure 2. Nuclear magneto-mechanical spectra obtained with RuO-thermometers. Dotted line shows the dipole field. The left spectrum comes from nondegenerate populations and the right one – from degenerated ones in low field. Left and right numbers in percents show polarizations before and after cross-relaxation, correspondingly. All spectral lines appear only during cross-relaxation and sweep field.

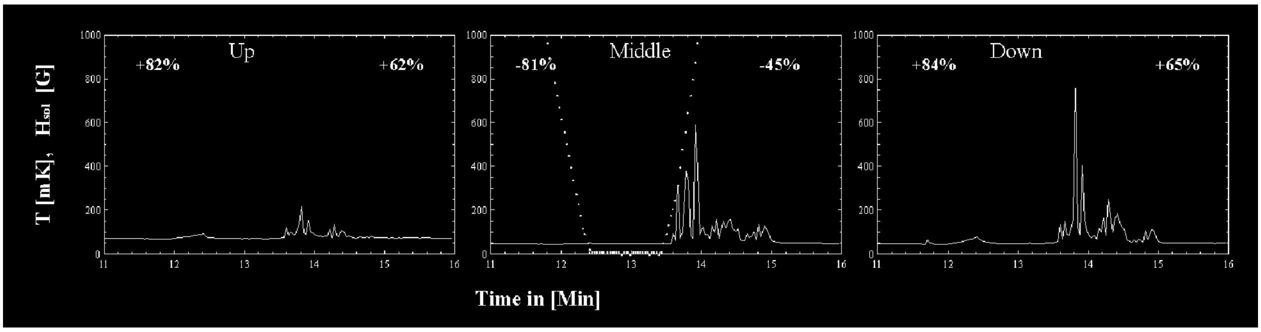

Figure 3. Magneto-mechanical spectra of high resolution obtained in the solenoid field (dotted line). The field was zeroed for 1 min to check the absence of a relaxation through electron impurities. Thermal pulses have the characteristic durations of the order of 10 sec in good agreement with our estimations made in Section 2.2.

of a high resolution obtained in homogeneous solenoid field. One can see, the thermal pulses have typical duration of the order of ten seconds that is in good agreement with the estimation for the relaxation time during the proton-nitrogen cross-relaxation, done in the Section 2.2. This result support the assumption made by R. Pound [4] about the important role played quadruple nuclei in the spin-lattice relaxation process.

Fast relaxation, observed in our experiments, could play an important role in the technique of the frozen polarized targets. In fact, owing to non-secular terms in nuclear dipole-dipole interactions, a weak coupling with quadruple spins must also lead to shortened relaxation time of any kind negatively polarized spin species. This effect may influence also on the value of reachable DNP-polarization which should be higher for negatively polarized spins relatively to positive one due the shortened built-up time at the lowest temperatures.

3.2. *Summary*

1. Nuclear magneto-mechanical effect was observed in the irradiated ammonia at superlow temperatures and at negative nuclear polarizations. The effect consists in the transformation of nuclear magnetic moment in the mechanical vibrations of a lattice.
2. Nitrogen spins in NH_3 produce the lattice vibrations when their alignment is being varied at the proton-nitrogen cross-relaxation at about 0.03 T. The characteristic time duration of vibration was founded in good agreement with our estimations.
3. Lattice vibrations were recorded with RuO-thermometers; they are spread along liquid helium and they cause the fast nuclear relaxation also in the cells with positively polarized material.
4. Evidence was produced, that due to non-secular dipole-dipole interactions with quadrupolar nuclear spins, the relaxation times of all others spin species should become shorter at negative relatively to positive polarization in high magnetic fields.
5. By the same reason, one can expect the higher reachable negative polarization relatively to positive one due to its faster built-up time at lower lattice temperatures.

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