NUCLEAR MAGNETOMECHANICAL EFFECT AT NEGATIVE SPIN POLARIZATION

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Abstract

If the magnetic properties of electrons and nuclear spins are widely used in applications, to our knowledge, the mechanical resonance of nuclear spins has not been observed yet. Almost hundred years later the [E-de H] experiment with a freely suspended magnet [1] we are looking for a way to detect the nuclear magnetomechanical effect using Dynamic Nuclear Polarization method, the nuclear demagnetization at superlow temperature and the world largest polarized target at CERN.

1. Idea and Numeric estimations

"*The physical fact underlined all gyromagnetic effects is that the nuclear spin and the electron spin, as well as their orbital angular moments, generate a magnetic moment parallel to the angular momentum with a magnitude fixed through a characteristic constant ...*" [2]. The first magnetomechanical experiment by A. Einstein and W.J. de Haas [E-de H] was published in 1915 [1]. It was shown that a freely suspended magnet, placed in a solenoid field, has a mechanical resonant oscillations at a specific frequency of a weak magnetizing field. There was a proof that the law of conservation of electron orbital magnetic moment allows "*...the occurrence of compensating angular momentum of another kind; the latter will be a crude mechanical angular moment*" [1].

The concept of "Nuclear Spin" was elaborated much later in 1925÷1927. Assuming the equal numbers of protons and orbital electrons in the samples, one can see that, in an analogical to [E-de H] experiment, nuclear spins should be cooled down from room temperature (≈ 300 K) to about $300\ \mathrm{K}/(\mu_e/\mu_P)^2 \approx 7 \cdot 10^{-4}$ K, where μ_e/μ_P is the ratio of electron to proton magnetic moments. This spin temperature could be reached in solid dielectrics in two steps: by Dynamic Nuclear Polarization (DNP) method and then by demagnetization of spins in reduced field. The problem comes from the relaxation of the spin energy which goes mainly through the interaction of nuclear spins with electron impurities because an amount of lattice phonons with nuclear frequencies is extremely small at low temperatures. In irradiated ammonia (NH_3) [3], investigated below, these relaxation time reaches thousands hours at 2.5 T and of the order of a few minutes at zero field and at about of 60 mK in both cases. Obviously, to identify the nuclear magnetomechanical effect, the nuclear spin-lattice relaxation must be faster than relaxation caused by electron impurities to avoid ambiguous interpretation of the effect.

R. Pound showed [4] that, under certain conditions, the relaxation rate of quadrupole nuclei interacting with an electric field gradient can largely exceed the relaxation rate through paramagnetic impurities. If it is so in ammonia at superlow temperatures, then the quadrupole nitrogen spins (I_N=1) could generate mechanical vibrations of a lattice. The effect may be enhanced during the cross-relaxation between proton and nitrogen spins because the proton moment (μ_P), and the energy, are an order of magnitude larger than for nitrogen (μ_N). The comparison of relaxation times of polarized proton through impurities and quadrupole spins can be estimated as follows [4]. The energy of dipolar interactions (E_{d-d}) between spins in NH$_3$ at a distance $\langle r_{PN}\rangle_{d-d}$ equals to

$$E_{d-d} = \frac{\mu_P \cdot \mu_N}{\langle r^3_{PN}\rangle_{d-d}} \approx \frac{1.4 \cdot 10^{-23} \cdot 2.0 \cdot 10^{-24}}{\langle r^3_{PN}\rangle_{d-d}} \approx \frac{3.0 \cdot 10^{-47}}{\langle r^3_{PN}\rangle_{d-d}}\,, \quad (c.g.s.) \tag{1}$$

which is considerably less than the interaction energy (W_q) between nitrogen quadrupole moment (eQ) and the averaged gradient of electric field in a lattice [5]

$$W_q = \frac{e^2 Q}{4 \cdot \langle r^3_N\rangle_q} \langle (3\cos^2\theta - 1)^2 \rangle\rangle_\theta \approx \frac{(4.8 \cdot 10^{-10})^2 \cdot 2.0 \cdot 10^{-26}}{4 \cdot \langle r^3_N\rangle_q} \cdot \frac{4}{5} \approx \frac{9.0 \cdot 10^{-46}}{\langle r^3_N\rangle_q}\,, \tag{2}$$

where $\langle r_N\rangle_q$ is the average distance between nucleus and electron charges (e), θ is the angle between the principal axis of the field gradient tensor and the direction of the magnetic field. Assuming that in the same material $\langle r^3_{PN}\rangle_{d-d} \approx \langle r^3_N\rangle_q$ we obtain the ratio of quadrupole to dipolar relaxation times T_q/T_{d-d} as [4]

$$\frac{T_q}{T_{d-d}} \approx (E_{d-d}/W_q)^2 \approx (3.0 \cdot 10^{-47}/9.0 \cdot 10^{-46})^2 \approx 0.001\,, \tag{3}$$

If, for example, the relaxation time through the electron impurities is of about 1 hour, as it is in irradiated ammonia at about of 0.03 T and 60 mK, then the quadrupole relaxation from Eq. 3 yields the seconds. We use this feature of quadrupole relaxation to extract the nuclear magnetomechanical effect from relaxation through the electron impurities.

The conversion of nuclear moment into lattice vibrations must change both the energy $\langle H_q\rangle$ and the alignment $A(N)$ of nitrogen spin system. At a low magnetic field we have

$$\langle H_q\rangle = h\nu_q(3\cos^2\theta - 1)\langle 3{I_z}^2 - I(I+1)\rangle = h\nu_q(3\cos^2\theta - 1)A(N)\,, \tag{4}$$

where h is 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 is the spin and $\langle I_z\rangle$ is z-component of angular moment. It is clear from Eq. 4 that the lattice vibrations should come from varying angular moment $\langle I_z\rangle$ in alignment.

2. Experimental Results

The data were obtained with the Compass polarized target at CERN. It uses a powerful dilution refrigerator, a solenoid with a homogeneous longitudinal field of 2.5±4 $\cdot 10^{-5}$ T and a dipole magnet producing a transverse field of 0.6 T. The dilution chamber consists of three cells (30+60+30) cm long and 4 cm in diameter (see Fig. 1), filled with irradiated and granulated ammonia [3]. Microwave (MW) cavity is also subdivided in three cells made of copper and electrically isolated from each other by MW-stoppers. It operates in

λ=4 mm wavelength range. Operating temperatures range is from 0.06 to 0.25 K. The nuclear polarization is measured by ten commercial "Liverpool" Q-meters connected to probing coils equally distributed along the target material [3]. The receiver circuits were permanently tuned to ν_0=106.42 MHz and fed by a RF-synthesizer. The frequency was scanned by 1000 steps within 600 kHz bandwidth.

Detailed investigation of the cross-relaxation in the ammonia at positive polarizations was done in [3], 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 studied the line shape of both nuclear species but the relaxation processes were not considered.

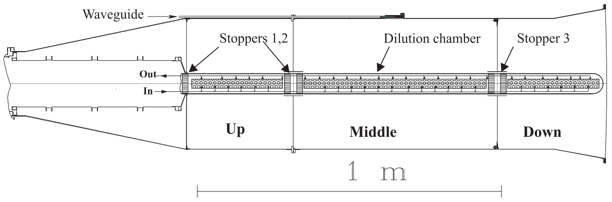

Figure 1: Up, Middle and Down microwave cells, powered through the waveguides. Electrical isolation between cells is performed with Stoppers 1,2,3; its design allows the free helium flow.

In this study, at field of 2.5 T, the proton spins in ammonia were polarized by the DNP method to ±80% then tests were performed in frozen mode 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 did not affect thermometers located near the ammonia. This means that at positive polarization the nuclear cross-relaxation is an adiabatic process which goes without any visible lattice effects. The cross-relaxation produces only a partial exchange between polarizations of spin species.

Nuclear magnetomechanical effect was only observed at negative polarizations. Fig. 2 shows thermal spectra recorded during double sweep through cross-relaxation field nearby

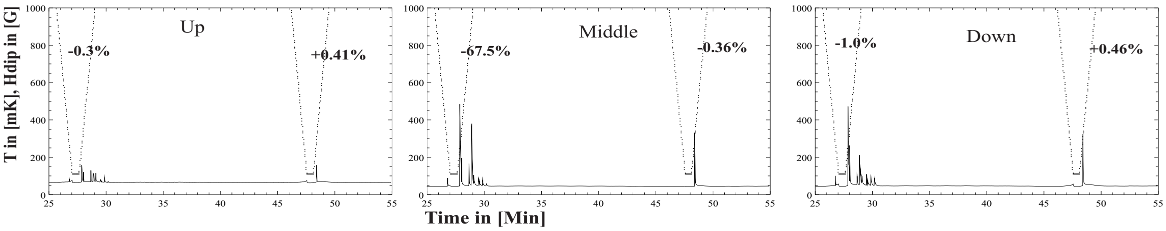

Figure 2: Magnetomechanical spectra obtained with RuO-thermometers. Dotted line shows the dipole field. Left and right percentages show the proton polarization before and after cross-relaxation. All spectral lines appear only during cross-relaxation and during the sweeping field.

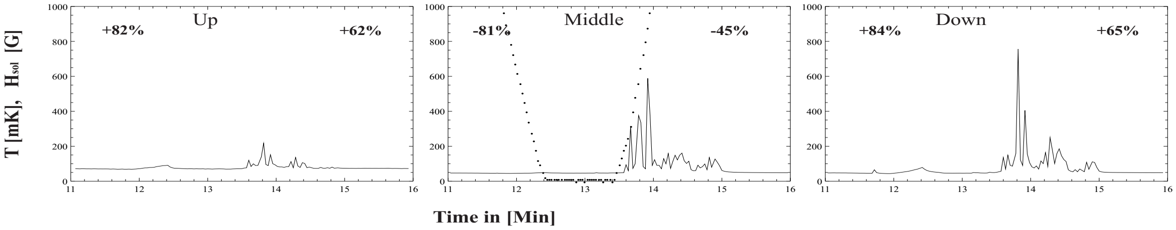

Figure 3: Magnetomechanical spectra of high resolution obtained in the uniform solenoid field (dotted line). Thermal pulses have characteristical durations of the order of 10 seconds in good agreement with estimation from Eq.3. This time the magnetic field went to zero for a minute to demonstrate an absence of relaxation through electron impurities during this short exposure.

0.03 T. Since cells are electrically isolated from each other, we conclude that the obtained spectra comes from the mechanical vibrations of the lattice. Such the explanation also confirms and almost identical shapes of spectral lines in different cells, seen in Fig. 2; it is clear that vibrations are generated in the Middle-cell which had the highest and negative initial polarizations and then excitations are spread to the nearby cells due to the free helium flow through Stoppers (see Fig. 1). Fig. 3 shows the high resolution vibration spectra obtained in homogeneous solenoid field. Typical duration of relaxation processes is of the order of ten seconds which confirms the estimation from Eq. 3 and the proposition by Pound [4] about the important role of quadrupole nuclei in the spin lattice relaxation.

The fast relaxation observed here may clarify the shorter relaxation time of negatively polarized spin species when interacting with quadrupole nuclei through a weak dipolar interaction. Magnetomechanical effect may also influence the reachable polarization which is usually higher at negative than at positive polarization, if the material contains quadrupole nuclei. In this case DNP-method can "polarize" at lower temperatures.

3. Conclusion

1. Nuclear magnetomechanical effect was observed in the ammonia at superlow temperatures and at negative nuclear polarizations. Effect consists in the transformation of nuclear magnetic moment into the mechanical vibration of the lattice.

2. Nitrogen spins in NH_3 produce the lattice vibrations when their alignment is varied during the proton-nitrogen cross-relaxatio at about of 0.03 T field.

3. Lattice vibrations were recorded with RuO-thermometers; they are spread along liquid helium and they cause the fast nuclear relaxation also in the nearby cells.

4. Evidence was produced, that, owing to a weak dipole-dipole coupling with quadruple spins, the magnetomechanical effect may produce a faster relaxation for negative compare to positive polarized spin species.

5. From the same reason, the negative reachable polarization should be higher than the positive one because, in this case, DNP-method can polarize at lower temperatures.

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