Rotating Transverse Helical Nuclear Magnetic Ordering

C. Urbina, J. F. Jacquinot, and M. Goldman
Service de Physique du Solide et de Résonance Magnétique, Centre d'Etudes Nucléaires de Saclay, F-91191 Gif-sur-Yvette, France

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Experimental evidence for the production of a nuclear dipolar magnetic ordering in a high external field, where the nuclear spins are oriented at right angle to the field in the form of a helix which precesses around the field at the Larmor frequency, is presented.

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The production of nuclear dipolar magnetic ordering in diamagnetic crystals, and its study by nuclear magnetic resonance or neutron diffraction, have been described in several publications.[1-4] Their main features are briefly recalled below.

The critical temperature T_c for the dipolar ordering of nuclear magnetic moments is in the microkelvin range, because of the smallness of these moments. It is only the nuclear spins that are cooled, by a two-step process: dynamic nuclear polarization in a high field H_0, followed by a nuclear adiatatic demagnetization in high field through suitable rf irradiation (adiabatic demagnetization in the rotating frame, in short ADRF). The spin temperature can be chosen positive or negative.

In a frame rotating around the direction Oz of the field H_0 at the Larmor frequency $-\gamma H_0$, the effective Hamiltonian governing the properties of the spin system is the part of the dipolar Hamiltonian which commutes with I_z. This truncated, or secular, dipolar Hamiltonian $\mathcal{H}_{II'}$ depends on the orientation of H_0 with respect to the crystal axes.

In all the cases we have studied up to now, the nuclear magnetic ordering, ferromagnetic or antiferromagnetic, was longitudinal, i.e., the spins were aligned along H_0, and the ordered structure looked the same in the rotating frame as in the laboratory frame. This is not so for a transverse ordering: When the spin orientations are at a right angle to H_0, it is only when viewed in the rotating frame that the ordered structure looks static. In the laboratory frame, the transverse spin polarizations are precessing around H_0 at the Larmor frequency, with constant magnitudes and *relative* orientations. A transverse nuclear magnetic ordered structure in high field is therefore a macroscopic coherent superposition of quantum spin states, which is stable in the absence of external couplings.

A hint for the actual existence of a transverse nuclear magnetic structure was suggested by susceptibility measurements in $Ca(OH)_2$.[4,5]

In this Letter, we describe experimental evidence for the production of a transverse ordered structure of the ^{19}F spins in CaF_2, after ADRF at $T > 0$ with $H_0 \| [111]$. For these conditions, the local Weiss-field approximation[1,4] predicts that two different orderings are degenerate: a longitudinal ferromagnet with domains whose dimension along H_0 is much larger than one of their transverse dimensions, and a transverse helix of wave vector $\vec{k}_0$ parallel to H_0 and small ($k_0 a \ll 1$, where a is the lattice parameter) but otherwise arbitrary. The bulk transverse magnetization vanishes for such a structure, and the spin system is not affected by radiation damping. On the other hand, the macroscopic detection of such a structure is not possible. In order to discriminate between these two orderings we have studied the rate of polarization of the spins of ^{43}Ca by off-resonance irradiation.

^{43}Ca, the only magnetic isotope of calcium, has an abundance of 0.13% and a gyromagnetic ratio 14 times less than that of ^{19}F. It does not perturb significantly the fluorine spin system, and is used as a microscopic probe of its ordering. Under rf irradiation of frequency ω at a distance Δ from the ^{43}Ca Larmor frequency, these spins are thermally coupled to the ^{19}F spins and become polarized if the latter are cold.

The polarization of rare spins S (^{43}Ca) by rf thermal mixing with abundant spins I (^{19}F) has been extensively studied at high spin temperature.[6,7] It exhibits markedly different features in two limiting cases, whose brief description is necessary to understand the present experiments.

(i) The spins I are cooled by an ADRF. In a frame rotating at the Larmor frequency ω_I for the spins I and at the frequency ω for the spins S,

the effective Hamiltonian of the system is

$$\mathcal{H} = \mathcal{H}_{II'} + \mathcal{H}_{IS'} + \Delta S_z + \omega_1{}^S S_x$$
$$= \mathcal{H}_{II'} + \mathcal{H}_{IS'} + \omega_e S_Z, \quad (1)$$

where $\omega_1{}^S = -\gamma_S H_1{}^S$ is the Larmor frequency in the field $H_1{}^S$ of irradiation of spin S. The effective field experienced by the spins S is along a direction OZ which makes an angle $\theta = \tan^{-1}(\omega_1{}^S/\Delta)$ with the direction Oz of H_0. Their corresponding Larmor frequency is

$$\omega_e = (\Delta^2 + \omega_1{}^{S2})^{1/2} \simeq \Delta \quad \text{if } \omega_1{}^S \ll \Delta.$$

The secular dipolar coupling $\mathcal{H}_{IS'}$ is of the form

$$\mathcal{H}_{IS'} = \sum_{i,\mu} B_{i\mu} I_z{}^i S_z{}^\mu \ll \mathcal{H}_{II'}. \quad (2)$$

The coupling $\mathcal{H}_{SS'}$ between spins S is negligible. $\mathcal{H}_{IS'}$ contains a part

$$V = \sin\theta \sum_{i\mu} B_{i\mu} I_z{}^i S_X{}^\mu \quad (3)$$

which commutes neither with $\mathcal{H}_{II'}$ nor with $\omega_e S_z$, and produces a thermal coupling between these reservoirs. The energy-conserving elementary processes of the thermal mixing are a flip of a spin S along its effective field together with a transition between two dipolar states of the spins I with the same value of I_z.

The polarization rate is of the form

$$W(\omega_e) = \sin^2\theta f(\omega_e), \quad (4)$$

where the rate factor $f(\omega_e)$ reflects the frequency spectrum of dipolar transitions among spins I. It is a monotically decreasing function of ω_e.

(ii) A large rf field is applied at the Larmor frequency of the spins I and their polarization is locked along its direction. The effective Hamiltonian in the doubly rotating frame is now

$$\mathcal{H} = \omega_1{}^I I_x + \mathcal{H}_{II'} + \mathcal{H}_{IS'} + \omega_e S_Z, \quad (5)$$

where $\omega_1{}^I$ is the Larmor frequency of the spins I in the large rf field. Since the spins I are quantized at right angle to Oz, the only energy-conserving elementary processes produced by the coupling V [Eq. (3)] are a flip of a spin S along the direction OZ of its effective field together with a flip of a spin I along the direction Ox of its effective field. The polarization rate of the spins S is still of the form (4), but the rate factor $f(\omega_e)$ is maximum when the effective frequencies of the two spins species are equal: $\omega_e = \omega_1{}^I$.

Let us now consider the case when, following an ADRF of the highly polarized spins I, these spins are in a dipolar ordered state. The transitions produced by the coupling V will be different depending on whether the ordering of the spins I is longitudinal or transverse.

If the ordering is longitudinal, V cannot produce the flip of a single spin I. The allowed thermal mixing transitions are of the same nature as those occurring in the high-temperature ADRF case. By similarity with this case, the rate factor $f(\omega_e)$ is expected to be a monotonic decreasing function of ω_e. This was checked by measuring the rate of polarization by off-resonance irradiation of ^{43}Ca, after an ADRF of the ^{19}F spins of CaF_2 at $T < 0$ and with $H_0 \| [111]$. There is a large body of theoretical and experimental evidence that under these conditions the ordered state is a longitudinal ferromagnet with domains.[4,8] The variation with ω_e of the rate factor $f(\omega_e)$ was indeed very close to that observed at high spin temperature.

If the ordering of the spins I is transverse, each spin I_i experiences a rotating transverse dipolar field from its neighbors, which plays the same role as the transverse *external* rf field in the high-temperature spin-lock case. One then expects that the rate factor $f(\omega_e)$ for the polarization of the spins S will be maximum when $|\omega_e|$ is equal to the Larmor frequency of the spins I in their Weiss field: $|\omega_e| = |qp|$, where q is the Weiss-field factor of the spins I and p their polarization.

We report the results obtained after an ADRF of ^{19}F in CaF_2, for $T > 0$ and $H_0 \| [111]$, starting from an initial polarization $p_i \simeq 0.80$. The polarization rate of ^{43}Ca under irradiation with an rf field $H_1 = 3.5$ G was measured at different irradiation offset fields. The rate factor $f(\omega_e)$ for this polarization is plotted in Fig. 1 as a function of the ^{43}Ca effective field. For comparison, the variation of $f(\omega_e)$ observed at high spin temperature[7] is also plotted in the figure.

The difference between the two curves is striking. In the present experiment, $f(\omega_e)$ is sharply peaked around an effective field $H_{e_0} \simeq 27$ G, corresponding to $\omega_{e_0}/2\pi \simeq 7.7$ kHz.

The variation of $f(\omega_e)$, of the spin-lock type, is as expected for a transverse structure and contrasts strongly with the behavior observed with a longitudinal ferromagnet.

For the theoretical transverse helix, the Weiss-field factor is

$$q = -\frac{4\pi}{3}\frac{\gamma_I{}^2\hbar}{2a^3}; \quad q/2\pi = -11 \text{ kHz in } CaF_2.$$

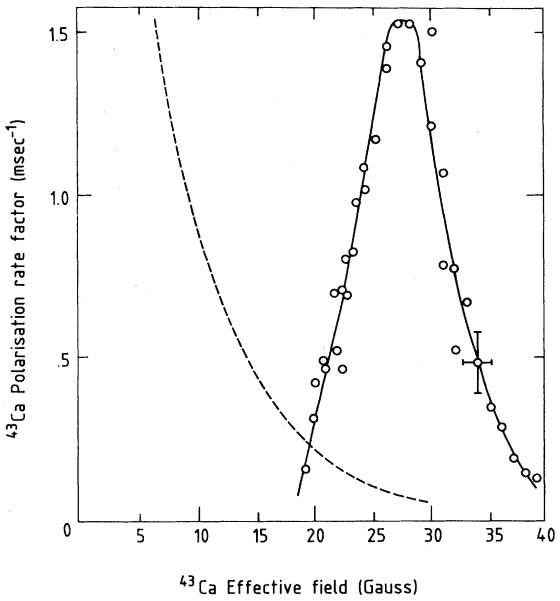

FIG. 1. Polarization rate factor of ^{43}Ca against its effective field in CaF_2 with $H_0 \| [111]$, after ADRF of ^{19}F spins at $T > 0$ with initial polarization $p_i \simeq 0.8$. rf field on ^{43}Ca: $H_1 \simeq 3.5$ G. Broken curve: high-temperature results in ADRF case (Ref. 7). $\gamma(^{43}\mathrm{Ca})/2\pi = 0.2865$ kHz/G.

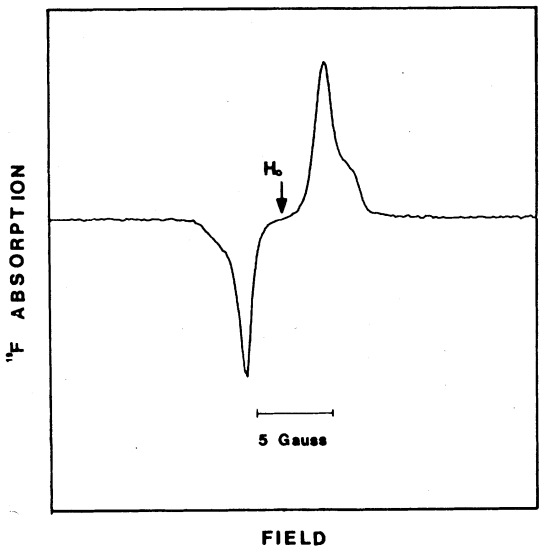

FIG. 2. Resonance signal of ^{19}F in CaF_2 with $H_0 \| [111]$, after ADRF at $T > 0$ with initial polarization $p_i \simeq 0.8$.

Within the simple model used above, the value $\omega_{e_0}/2\pi = 7.7$ kHz for which $f(\omega_e)$ is maximum corresponds to a transverse ^{19}F polarization $p \simeq 0.7$, that is, according to the Weiss-field approximation, to a temperature $T \simeq 0.8 T_c$, where T_c is the critical temperature, of theoretical value $T_c = -q/2 = 0.268$ μK.

In a more sophisticated description of the thermal mixing, the coupling V of the spins I with the inhomogeneous dipolar field of the spins S, normal to the axes of quantization of the spins I, is able to create or annihilate elementary excitations of wave vector $\vec{k}$ and frequency $\omega(\vec{k}) = \omega_e$. The simplest temperature-dependent approximation for elementary excitations, namely, the random-phase approximation, predicts for $H_0 \| [111]$ an excitation spectrum which is narrow and peaked around qp. The experimental variation of $f(\omega_e)$, in Fig. 1, is about twice as broad as predicted by this approximation, which may be interpreted as arising from the finite lifetime of the elementary excitations.

When the fluorine system is allowed to warm up by the spin-lattice relaxation, the peak of $f(\omega_e)$ is shifted to lower values of ω_e, because of the decrease of the ^{19}F transverse polarization. At the same time the function $f(\omega_e)$ is increasingly broadened, because of the shortening of the elementary excitation lifetime.

As a further check of our interpretation, we have compared the polarization rate factor of Fig. 1 with the absorption signal of the ^{19}F spins.

In the standard Weiss-field approximation, the resonance frequency of the helical structure is

$$\Omega_{res} = (\tfrac{3}{2})^{1/2} qp \simeq 1.225 qp. \quad (6)$$

The resonance signal of ^{19}F is shown in Fig. 2. Assuming the distance between the two peaks to be $2\Omega_{res}$, one finds

$$\Omega_{res}/\omega_{e_0} \simeq 1.06$$

which is resonably close to the prediction of Eq. (6), because of the uncertainties in the theory. The bumps on the wings of the resonance signal are accounted for as due to the simultaneous creation or annihilation of two elementary excitations by the rf field of observation. The theory of this effect will be given in a future article.

The experimental facts—existence of a finite effective frequency ω_{e_0} at which $f(\omega_e)$ is maximum, contrasting with the observations on a longitudinal ferromagnet; narrow width of the function $f(\omega_e)$; and ratio of ω_{e_0} to the fluorine resonance frequency—provide a convincing evidence that the ordered structure under study is indeed a rotating transverse helical structure.

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Does the Standard Hot-Big-Bang Model Explain the Primordial Abundances of Helium and Deuterium?

N. C. Rana
Tata Institute of Fundamental Research, Bombay 400 005, India
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It is shown that according to the standard hot-big-bang nucleosynthesis calculations, no single value of the ratio η of baryon to photon number density can explain the right abundances of both ^{4}He and ^{2}D. A slight neutrino degeneracy, however, provides a testable solution for η which is in conformity with the current estimate of the baryonic component of the density parameter $(\Omega_N)_0$ of the Universe.

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A major success of the standard hot-big-bang model of the Universe is claimed to be that it provides a satisfactory explanation for the universal abundance of helium and deuterium.[1] Recently Stecker[2] has pointed out some inconsistencies between the theoretical predictions and observations of the abundances of these two elements which take into account the present estimates of the Hubble constant H_0, the baryonic component of the density parameter $(\Omega_N)_0$, and the temperature of the microwave background radiation $(T_\gamma)_0$. It is shown here that this inconsistency exists at a more basic level than implied by Stecker's arguments.

The theoretical calculations of the abundances of ^{4}He and ^{2}D depend essentially on three parameters, the ratio (η) of baryon to photon number density during the time of nucleosynthesis, the neutron half-life $(\tau_{1/2})$, and the effective number of neutrino families (N_ν) contributing to the total density. N_ν is generally taken to be an integer ≥ 3.[2] Various laboratory measurements on $\tau_{1/2}$ have not so far produced a conclusive result, although three values (namely $\tau_{1/2} = 10.13$, 10.61, and 19.82 min) chosen by Olive *et al.*[1] could represent its possible range of uncertainty. I adopt the same values for the present analysis.

Five independent, up-to-date, observational estimates of the primordial abundance of ^{4}He by mass (Y_p) have been quoted by Olive *et al.*[1] and I take the statistical mean of these estimates, which is $(Y_p)_o = 0.220 \pm 0.008$. The observational estimate[3] of the abundance of ^{2}D by mass may be taken as $(X^D)_o = (3.6 \pm 0.8) \times 10^{-5}$. Note that subscript o refers to the observational estimates whereas p refers to the primordial value. The primordial abundance of deuterium is, however, not known. But it should not be very much different from the above value for the following reason. ^{2}D can be both synthesized and destroyed only through the stellar type of nucleosynthesis which results in a very little balance of ^{2}D in the long run. If the entire ^{2}D abundance is not of primordial origin, it would be very difficult to give an astrophysical explanation for its origin. So, $(X^D)_p \gtrsim (X^D)_o$. Again, if we assume that $Y_p < 0.228$, the balance of the net universal abundance of ^{4}He (which is about 0.05–0.08 by mass) must have been produced through the stellar type of nucleosynthesis. This implies that only about 10%–12% of the total baryonic matter has ever been processed through stellar cycles. This pro-

