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ROTARY SPIN ECHOES

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Torrey[1] has observed the free precession of nuclear spins around an rf field H_1, fixed in a frame rotating at the Larmor frequency $\omega_0 = \gamma H_0$ around a large dc magnetic field H_0. He showed that, for an H_1 much larger than the inhomogeneity of H_0, the latter has a negligible effect on the decay of the spin magnetization which is mainly due to the inhomogeneity of H_1. We report here on a method of overcoming the inhomogeneity of H_1 by the production of echoes in the rotating frame ("rotary echoes") which are very similar to the usual spin echoes.[2] The rotary echoes, however, have some additional specific features that make them particularly suitable for the measurement of long relaxation times.

Consider the rotating frame with the z-axis along H_0 and the x-axis along the rf field H_1. If at the time $t=0$ we suppose that the spin magnetization M is along the z-axis, M will precess in the yz plane and, at $t=\tau$, the angle of precession is $\phi = \gamma H_1 \tau$. In practice H_1 is inhomogeneous, with a half-width ΔH_1, and will vary within the sample. Therefore, the total magnetization, sum of magnetization vectors at different points of the sample, will fan out in the yz plane in a time of the order of $1/\gamma \Delta H_1$.

At $t=\tau$, we perform a 180° phase shift on the rf field so that H_1 is suddenly reversed in the rotating frame. From then on, the magnetization vector, at each point, will precess at the same rate as before, but in the opposite direction. Therefore, at time $t=2\tau$, the angle of precession will cancel out and all the magnetization vectors will be in phase again on the z-axis producing an "echo," as shown in Fig. 1.

In that way, we overcome the effect of the inhomogeneity of H_1 and we measure only the effect of spin relaxation. For example, in a liquid where the Bloch equations[3] are valid, the envelope of the echoes, when τ is varied, is an exponential curve with a time constant T given

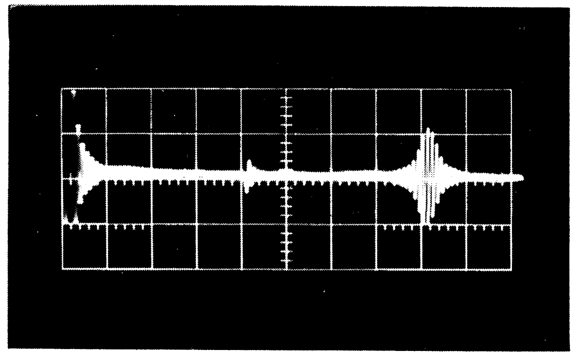

FIG. 1. A "rotary echo" in doped water. The total trace is 100 msec long.

by[1]

$$\frac{1}{T} = \frac{1}{2}\left(\frac{1}{T_1} + \frac{1}{T_2}\right),$$

where T_1 and T_2 are the longitudinal and transverse relaxation times, respectively. The main limitation of the rotary echo method is due to self-diffusion in the liquid just as in the standard spin-echo experiment. Its effect can be greatly reduced, as in the Carr-Purcell[4] method, by reversing H_1 (which corresponds to a 180° pulse in a spin-echo experiment) not only at $t=\tau$ but also at $t=3\tau$, $t=5\tau, \ldots$, $t=(2n+1)\tau$; the echoes then occurring at $t=2\tau$, $t=4\tau, \ldots$, $t=2(n+1)\tau$.

The main difficulty in applying the spin-echo method in the measurement of long relaxation times is that (a) in order to overcome diffusion a large number of 180° pulses must be applied, and (b) the errors on the 180° pulses being cumulative, a large number of pulses give poor reproductibility. A remarkable feature of the "rotary echoes" is that the errors on the reversals of H_1 are not cumulative. This can be seen by a detailed analysis in the rotating frame in a manner analogous to that described by Meiboom and Gill[5] for their modified Carr-Purcell method. The result is the possibility of obtaining a large number of echoes (1000 are shown in Fig. 2) with good stability, while it was practically impossible to obtain in the same magnet more than 30 echoes of the Carr-Purcell type. Figure 2 shows a measurement of relaxation time in oxygen-free benzene which yields the value $T_2 = 19.6 \pm 1.5$ sec.

We use a crossed coil spectrometer. The transmitting coil is fed by a gated oscillator giving an rf field H_1 up to 1.5 gauss (rotating component). As in Torrey's experiment,[1] we

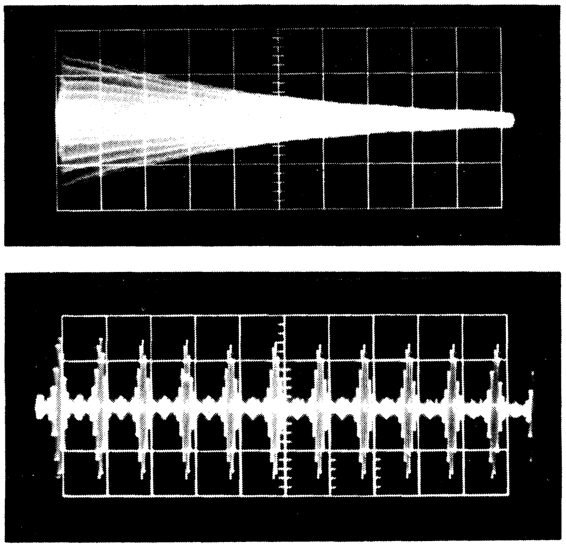

FIG. 2. Rotary echoes of the Carr-Purcell type in oxygen-free benzene. Upper photograph: the whole pattern. The pulse rate is 50 per second, and the total trace is 40 sec long. The rotary echoes are too close to be resolved on the photograph. Lower photograph: part of the same pattern enlarged 100 times to show the detail of the echoes.

turn on the oscillator at $t=0$, and the magnetization M precesses around H_1 in the yz plane. The receiving coil is tuned in the absorption mode, so that the audio signal observed after detection is directly proportional to M_y. Since the magnetization M precesses around the x-axis at the frequency γH_1, the frequency of the audio signal on the cathode ray oscilloscope gives the magnitude of H_1, while its decay gives the inhomogeneity of H_1. The reversal of H_1 at $t=\tau$ is obtained by the application on the grid of the oscillator of a strong pulse of variable length (1 to 20 microseconds). The frequency is then slightly changed for a short time and a phase shift occurs. This shift can be set to be 180°, either by direct observation of the rf voltage on a fast cathode ray oscilloscope, or by tuning the pulse length for maximum echo amplitude.

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PROPOSAL FOR A "STAIRCASE" MASER*

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Inversion by adiabatic fast passage of a single electron spin resonance transition in a multilevel system with unequal level spacings has recently been demonstrated.[1,2] Using this phenomenon, it appears feasible to construct a "staircase" maser which will radiate at approximately twice the pump frequency. Consider the three-level system of Fig. 1. If the 1↔2 transition is

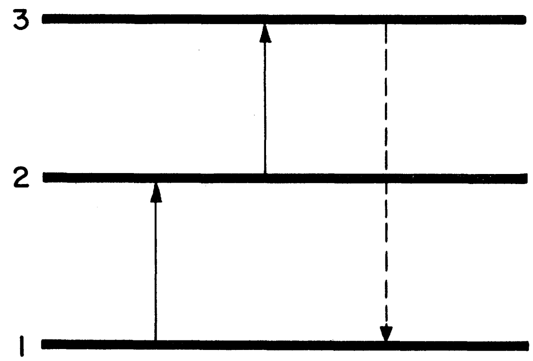

FIG. 1. In the staircase maser scheme, the 1↔2 and 2↔3 transitions are successively inverted by fast passage, followed by double-frequency amplification on the 1↔3 transition.

inverted by fast passage, followed in a time less than the spin-lattice relaxation time by inversion of the 2↔3 transition, a population inversion between levels 3 and 1 will result, permitting amplification on the 1↔3 transition at roughly double the pump frequency. Operating points can be found in common maser materials at which the two successive inversions can be performed by the same pump oscillator in a single sweep of the dc magnetic field. Extension of the method to more than two steps up a staircase is conceivable. Cross relaxation between the two nearly equal pump transitions may be the major difficulty in implementing this scheme.[3]

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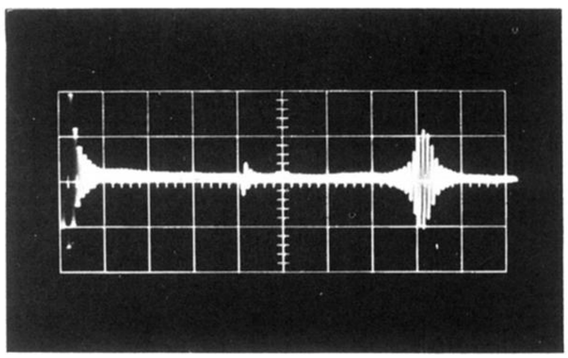

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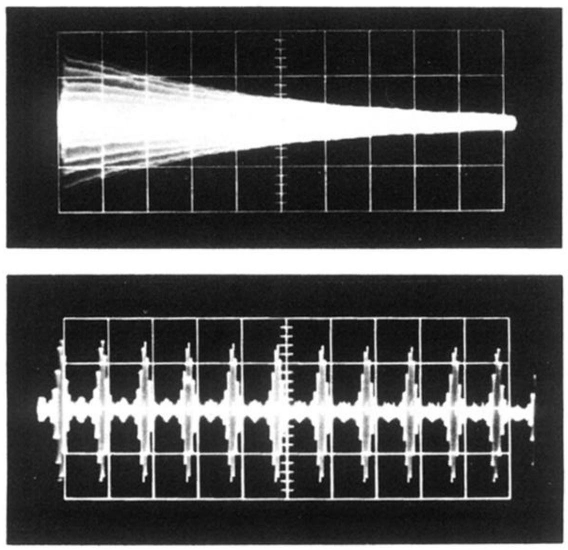

FIG. 2. Rotary echoes of the Carr-Purcell type in oxygen-free benzene. Upper photograph: the whole pattern. The pulse rate is 50 per second, and the total trace is 40 sec long. The rotary echoes are too close to be resolved on the photograph. Lower photograph: part of the same pattern enlarged 100 times to show the detail of the echoes.