# NUCLEAR SPIN ECHO IN THE ANTIFERROMAGNET $RbMnF_3$

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In the observation of nuclear magnetic resonance (NMR) in antiferromagnets having small anisotropy fields, nonlinear effects, connected with strong hyperfine interaction, may arise at low temperatures. Thus, for example, there is a dynamic NMR frequency shift (pulling, $\delta\omega_V$), which depends on the value of the nuclear magnetization [1 - 3]:

$$\frac{\omega}{\omega_n} = \left[1 - \frac{2\omega_E \omega_n \gamma_e m_z}{\omega_e^2 \gamma_n M_0}\right]^{1/2},$$

where $\omega$ is the NMR frequency, $\omega_n$ is the unshifted NMR frequency, $m_z$ is the nuclear magnetization, $\omega_E$ is the exchange energy, $\omega_e$ the AFMR frequency, and $M_0$ the electronic sublattice magnetization. In such antiferromagnets, new effects are possible in the observation of nuclear spin echo, since the echo signal is produced by applying to the sample sufficiently strong radio pulses, and appreciable changes occur in the longitudinal component of the nuclear magnetization $m_z$. Until now, however, the nuclear echo phenomenon was not investigated in these crystals.

We report here observation of echo of $Mn^{55}$ nuclei in $RbMnF_3$, a cubic antiferromagnet having small anisotropy fields (∿1 Oe), in which the NMR was investigated in sufficient detail by the usual stationary methods [4 - 6]. The $RbMnF_3$ single crystals were synthesized by the Stockbarger method in a graphite crucible in an atmosphere of purified argon. An atmosphere of elemental fluorine was used to purify the initial charge. The measurements were carried out in both polycrystals and single crystals, at liquid-helium temperature, using a spectrometer described in [7].

The nuclear echo was observed in the frequency range $600 < f_0 \leq 672$ MHz, with the external field $H_0$ ranging from 2.5 to 9 kOe (Fig. 1). The echo-signal intensity had an anomalous dependence on the applied external field. The intensity was minimal at $f_0 \simeq 610 - 620$ MHz ($H_0 \simeq 2.5 - 3$ kOe), and then increased by 10 - 100 times, reaching a maximum at $f_0 = 650$ MHz ($H_0 > 5$ kOe, signal/noise ratio ∿200), after which it decreased somewhat with increasing frequency ($H_0 > 5$ kOe). At frequencies $f_0 \simeq 650$ MHz, when the two-pulse procedure was used, we observed besides the usual echo also an additional echo (signal/noise ∿10), lagging the first by a time equal to the delay between the first and second pulses. We propose that the secondary echo is due to the fact that the first echo signal acts on the sample like an additional third radio-frequency pulse. The stimulated echo observed in the three-pulse measurement procedure also exhibits an unusual dependence of the intensity on $H_0$. The stimulated echo has a high intensity at $H_0 \simeq 7 - 8$ kOe, and is not observed in weaker external magnetic fields.

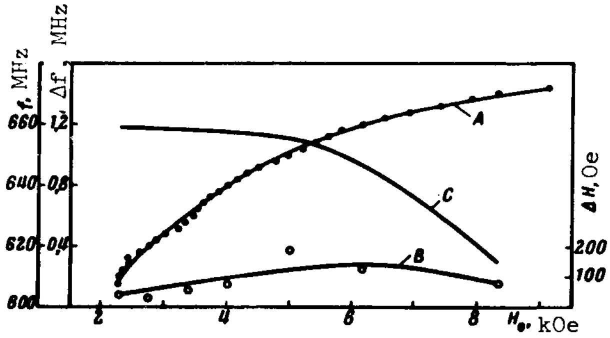

Fig. 1. Dependence of NMR frequency (A), line width ΔH (B), and Δf (C) on the external magnetic field $H_0$.

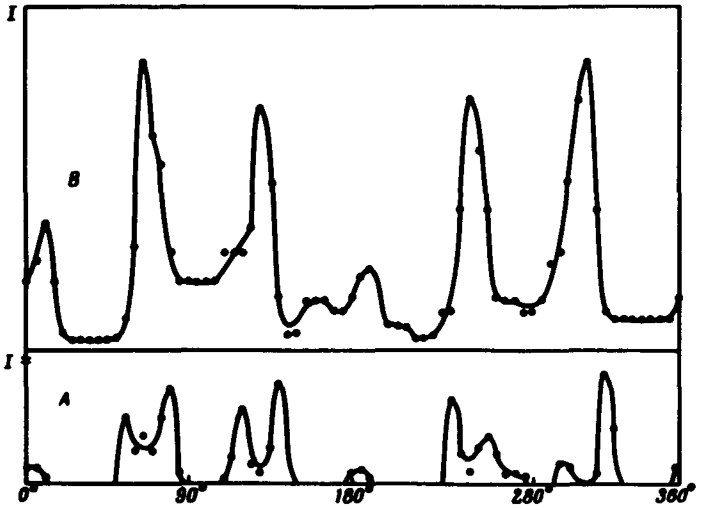

Fig. 2. Angular dependence of the echo intensity (A - 620 MHz, B - 660 MHz). The abscissas represent the angle between $H_0$ and [100]; the alternating field is parallel to [001].

The observed behavior of the echo intensity can be briefly interpreted as follows. In magnetically-ordered crystals, the signal intensity of the ordinary echo is maximal when the gain $\eta$ is maximal [3] and 120-degree pulses are used. In our case, for weak fields, $H_0 \approx 2 - 3$ kOe, the pulling is maximal. In principle it is impossible to rotate here the nuclear magnetization through 120°, for even at relatively small angles between the nuclear magnetization and the z axis the magnitude of the pulling changes and the radio-frequency pulse is no longer tuned to the new value of the resonant frequency. It turns out therefore that the larger the pulling, the smaller the echo signal, for in this case the nuclear-magnetization vector can be deflected only through small angles. With incerasing external constant field, the signal increases, since the pulling decreases, but then the signal begins to decrease, now as a result of the appreciable decrease of the gain $\eta$, since $\eta \approx H_{loc}/(H_0 + H_A)$ ($H_{loc}$ is the local field at the nucleus). According to our estimates, $\eta \approx 130$ at $f_0 \approx 630$ MHz.

The dependence of the intensity of the stimulated echo on the field $H_0$ is explained in similar fashion. The intensity of the stimulated echo is determined by the change of $m_z$ under the influence of the radio-frequency pulses. The magnitude of this change is small when the pulling is large, so that stimulated echo can be observed only in sufficiently strong external fields, where the pulling is decreased.

The variation of the resonant frequency as a function of the external field agrees with the data of [4]. Figure 1 shows also measurements of the NMR line width as a function of $H_0$, obtained in magnetic-field units. The line width was measured by varying the magnetic field. The calculated $\Delta f(H_0)$ curve, in frequency units, was obtained from the formula $\Delta f = (df_0/dH_0)\Delta H$, where the derivative $df_0/dH_0$ was determined from curve A (Fig. 1). The obtained $\Delta f(H_0)$ dependence agrees qualitatively with [6]. The measurements in [4, 6] were performed by stationary methods.

Figure 2 shows the dependence of the echo intensity on the sample orientation. In these measurements we used an $RbMnF_3$ single crystal of irregular shape. The sample was oriented in accordance with the characteristic cleavage on the surface. The field $H_0$ lies in the (001) plane and the alternating field is directed along the [001] axis.

The angular dependence of the intensity reveals a pronounced periodicity, as should be the case for a cubic crystal. A detailed analysis of this dependence, however, is rather complicated and will not be discussed in the present article.

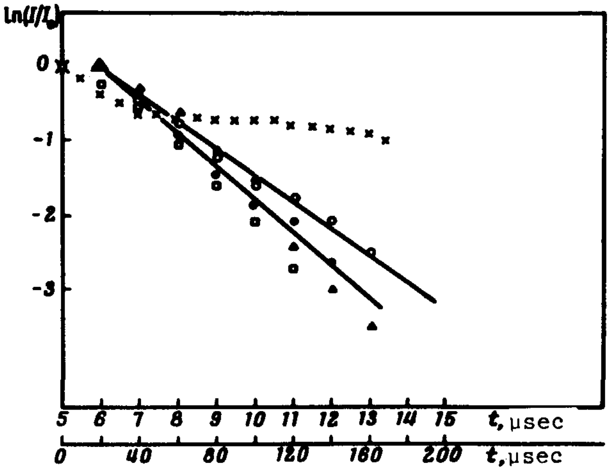

Fig. 3. Intensity of the signals of the ordinary echo (o - 620 MHz, o - 640 MHz, Δ - 666 MHz, □ - 670 MHz) and of stimulated echo (× - 670 MHz) vs. the delay time between pulses.

Figure 3 shows the variation of the signal intensity of the ordinary echo, and also of the stimulated echo, when the delay time is varied. As seen from the figure, in most cases the dependence of the echo signal intensity on the delay time is not exponential. The different character of the decrease of the ordinary-echo intensity (transverse relaxation) as a function of the resonant frequency (Fig. 3) can apparently be connected with the influence of the Suhl-Nakamura interaction (SNI) [8]. When the contribution of the SNI to the NMR line width is significant, Bloch's equations do not hold for the transverse relaxation, which is no longer exponential (666 and 670 MHz, Fig. 3). On the other hand, if the line width is determined by the inhomogeneous broadening, the relaxation can be exponential. The data shown in Fig. 3 indicate that the dependence is exponential where the line width is maximal, i.e., at frequencies 620 - 640 MHz. It is indeed known from the data of [6] that in this frequency region the NMR line is greatly and inhomogeneously broadened. A more correct analysis requires, however, that the spin-diffusion effects be taken into account [9].

The behavior of the stimulated echo can be represented in the form of a superposition of two exponentials (Fig. 3), with characteristic parameters $T_{1(1)} \approx 60$ μsec and $T_{1(2)} \approx 420$ μsec. The nature of such a behavior of the longitudinal relaxation is as yet unknown.

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