NMR in ferromagnetic Fe_2MnO_4 at millidegree temperatures

B. S. Dumesh

Institute of Physics Problems, USSR Academy of Sciences
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The spin-echo method is used to investigate NMR in Mn^{55} nuclei at temperatures 4–0.04°K. A large dynamic shift of the NMR frequency is observed for the first time in ferromagnets. The temperature dependence of the magnetization of Mn^{55} nuclei in the region of appreciable deviations from the Curie law is investigated by measuring the dynamic frequency shift.

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This paper is devoted to an investigation of NMR in Mn^{55} nuclei in the spinel ferrite Fe_2MnO_4 at temperatures 0.04—1 °K. Investigations in this temperature range solve two problems.

The first is a detailed study of the dynamic frequency shift (DFS) in ferromagnets.[1] For ferromagnets, the DFS is given by the formula

$$\delta f = f_{n0}\eta\frac{m}{M}, \tag{1}$$

where $f_{n0}=\gamma_n H_n$ is the hyperfine frequency, η is the gain, and m and M are the average magnetizations of the nuclei and the electrons. In our temperature range $M=M_0=\text{const}$ and

$$m = Ng_n\beta_n I B_I\left(\frac{g_n\beta_n I H_n}{kT}\right) \tag{2}$$

where β_n is the nuclear magneton, I is the spin of the nucleus, k is Boltzmann's constant, and B_I is the Brillouin function. It follows from (1) and (2) that the frequency shift becomes appreciable only at $T<1$ °K. In ferromagnets, the dynamic frequency shift was observed only in Fe_2MnO_4 at $T=2$ °K by Heeger and Houston.[2] However, the smallness of the effect (the shift of the NMR line at this temperature was smaller than the line width) did not enable them to carry out detailed investigations.

Second, the dynamic frequency shift yields directly with the aid of (1), the temperature dependence of the nuclear magnetization, and the use of temperatures on the order of 0.05 °K makes it possible to obtain in this case a high degree of nuclear-spin polarization.[1)] We can therefore investigate the temperature dependence of the nuclear magnetization in the region of appreciable deviations from the Curie law.

Low temperatures were obtained by us with the aid of a cryostat with He^3 dissolved in He^4, similar to that described in[3]. The cryostat operated in the circulation regime down to 0.04 °K, and the temperature was maintained constant within 10%. The temperature was measured with a carbon-resistance thermometer manufactured at our Institute[4] and (at temperatures 1.5—0.5 °K)

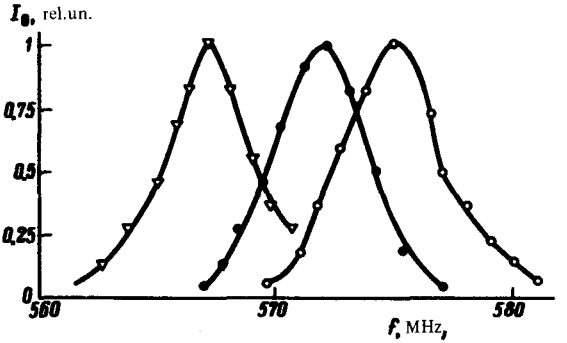

FIG. 1. NMR line in Fe_2MNO_4: ○—0.2 °K, ●—0.09 °K, ▽—0.04 °K.

with the aid of an He^3 condensation thermometer. To improve the thermal contact, the thermometer and samples were placed directly in the solution bath of the cryostat.

At temperatures $T \lesssim 0.1$ °K there is a strong increase in the Kapitza thermal resistance[5]

$$R = c/T^3, \tag{3}$$

where $c \approx 10^1$—10^2 cm² deg⁴/W. To prevent overheating the sample, it is necessary to dissipate in it not more than 10^{-3}—10^{-4} erg/sec. We therefore used for the measurements a spin-echo procedure (spin echo in Fe_2MnO_4 was first investigated in[6]). The pulsed NMR spectrometer used by us is described in[7]. In these experiments, the RF pulse power was $P \approx 10^{-3}$ W, the pulse duration $t_f \approx 3$—4 μsec, and the repetition frequency ≈ 1 Hz. These parameters enabled us to keep the samples from becoming overheated. The magnetic component of the RF field was directed along the [100] axis.

At $T \approx 4$ °K the NMR line has a flat top and a width $\Delta f \approx 10$ MHz. Application of an external magnetic field in the [100] direction reduces greatly the line

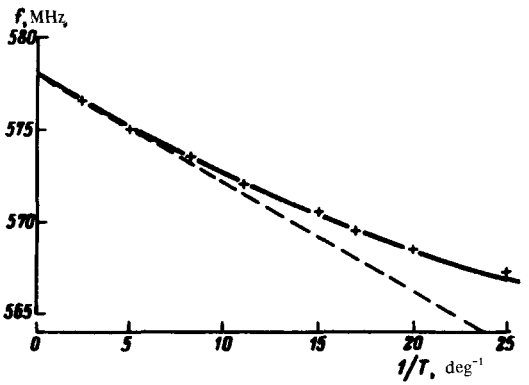

FIG. 2. Echo signal frequency vs. reciprocal temperature.

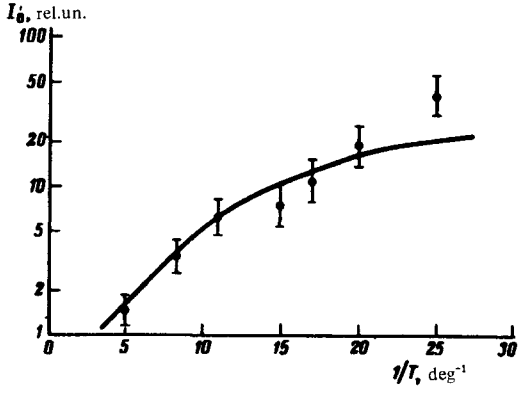

FIG. 3. Echo signal intensity vs. reciprocal temperature.

width and at $H \approx 5$ kOe we have $\Delta f \approx 3$ MHz. The value of H_n was determined by extrapolating f_{n0} to a zero external field. We obtained $H_n = 550$ kOe. The transverse relaxation time was $T_2 \approx 15$ μsec ($T = 4.2$ °K). Investigations at $T < 1$ °K were carried out without an external magnetic field. With decreasing temperature, the NMR line width decreases greatly and the line shifts to lower frequencies (Fig. 1). The line shape is independent of the RF power in this case. The gain is therefore homogeneous over the line and we observe NMR from nuclei situated inside the domains. The shift of the NMR line frequency as a function of the reciprocal temperature is shown in Fig. 2. As follows from (1) and (2), this dependence should be described, apart from a constant factor, by a corresponding Brillouin function. We used for the calculation the constants $I = 5/2$, $\gamma_n = 1056$, and $H_n = 550$ kOe. The obtained theoretical dependence is shown in Fig. 2 by a solid line. The dashed line corresponds to the Curie law at the same values of the constants. The maximum degree of nuclear polarization obtained by us was 65%. From the coefficient of proportionality of $\delta f(T)$ and $m(T)$ we can obtain the value of the gain. It turned out that $\eta \approx 200$, which corresponds in order of magnitude to resonance with nuclei inside the domains. From the formula $\eta \approx H_n/H_a$, where H_a is the anisotropy field, we could determine H_a, which was found to equal ≈ 3 kOe, in reasonable agreement with the data by others.[2,6] It can therefore be stated that the dynamic frequency shift and the magnetic moment of the nuclei are described by formulas (1) and (2) up to a high degree of polarization.

Figure 3 shows the dependence of the echo-signal intensity on the reciprocal temperature. With decreasing temperature, the signal increases much more rapidly than the magnetic moment (as would be the case if the echo signal were produced by the Hahn mechanism). On the other hand, at small moment rotation angles the relation[2)] $I_{\text{echo}} \sim m\delta f \sim m^2$ is satisfied. The solid curve of Fig. 3, plotted in accord with this relation, is in qualitative agreement with the results.

At low temperatures, the transverse relaxation time T_2 is strongly uneven over the NMR line. At $T = 0.05$ °K, $T_2 = 60$ μsec ($f = 570$ MHz), $T_2 = 80$ μsec ($f = 568$ MHz, center of the NMR line), and $T_2 = 140$ μsec ($f = 566$ MHz) and continues to increase with decreasing frequency. This phenomenon can be explained completely. The low-wing of the NMR line is due in this case to resonance in the domain walls. Indeed, the frequency shift is proportional to the gain (1),

which is determined by the local susceptibility and reaches a large value in the domain walls. But the number of spin states per frequency interval $F(f)$ is much smaller in the domain walls, and, as shown in[8], $T_2 \sim [F(f)]^{-1}$. By investigating $T_2(f)$ in detail we can therefore find the connection between two important characteristics of domain walls, the susceptibility and the density of states.

The longitudinal-relaxation time was determined by us by measuring the dependence of the signal on the repetition frequency of pairs of pulses and it turned out that $T_1 \simeq 0.2$ sec at $T = 0.05$ °K. We assume that what is measured in this case is not the true spin-lattice relaxation time, but the time constant due to the Kapitza thermal jump $\tau = RC$ (C is the heat capacity). There are no data on the heat capacity of Fe_2MnO_4 at low temperatures, and we are unable to establish the nature of the measured T_1.

In conclusion, the author is sincerely grateful to A. S. Borovik-Romanov for constant guidance of the work, and also S. M. Elagin and V. A. Krutikhin for help with constructing the cryostat.

[1)]The hyperfine interaction energy in Fe_2MnO_4 turned out in this case to be of the order of 0.03 °K in temperature units.
[2)]See, e.g., [7] regarding the frequency mechanism of echo production.

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