NMR on Mn^{55} nuclei in easy-plane antiferromagnets $MnCO_3$ and $CsMnF_3$ in the millidegree temperature range

B. S. Dumesh

Institute of Physics Problems, USSR Academy of Sciences
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Pulsed methods were used to investigate NMR on Mn^{55} nuclei in the temperature range 1–0.04°K. The functional connection between the frequency shift and the magnetic field agrees with the theory of coupled oscillations of the electronic and nuclear moments. The average nuclear magnetization of the Mn^{55}, however, obtained from the frequency shifts, turns out to be much less than that determined for a system of non-interacting moments.

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The coupling between the nuclear and electronic spin sub-systems in antiferromagnets at low temperatures is so strong, that a restructuring of the free oscillations of the magnetic moments takes place (the so-called dynamic frequency shift-DFS). The natural frequencies are determined in this case from the equation[1] [1)]

$$(f^2 - f_{e0}^2)(f^2 - f_{n0}^2) - f^2 f_T^2 = 0, \qquad (1)$$

where f_{e0} and f_{n0} are the "undisplaced" AFMR and NMR frequencies, $f_T^2 = \gamma_e^2 2H_E A\langle m\rangle$, γ_e is the electron gyromagnetic ratio, H_E is the exchange field, A is the hyperfine-interaction constant, and $\langle m\rangle$ is the average nuclear moment. In many experiments[1–3] the agreement between the observed spectra and the theory is satisfactory, but all the investigations were carried out in a narrow temperature interval (4.2—1 °K). Our study was devoted to NMR spectra in the temperature range 1—0.04 °K, at a large expected nuclear polarization. We chose for the investigation the easy-plane antiferromagnets $MnCO_3$ and $CsMnF_3$. In these substances, the NMR dynamic frequency shift reaches a maximum value and has been sufficiently well investigated at helium temperatures.[2,3] All the necessary constants were obtained in these studies.

We obtained low temperatures with the aid of a cryostat in which He^3 was dissolved in He^4. The temperature was measured with a carbon resistance thermometer[2)] and (in the range 1.5—0.5 °K) with the aid of an He^3-condensation thermometer. To improve the thermal contact, the samples and the thermometers were placed directly in the dissolution bath. All the experiments were performed on single-crystal samples. The constant magnetic field was produced with a superconducting solenoid. The error in the measurements of the magnetic field was determined by the graduation accuracy and amounted to 1%. The constant and the RF magnetic fields were mutually perpendicular and were in the basal plane of the samples. The NMR signal was registered by observing the free precession following the short RF pulse or the spin-echo signal of the Mn^{55} nuclei. The observation procedure and the NMR spectrometer are described in[4].

At the magnetic fields investigated by us, the frequency range was $f_{e0} \gg f_{n0} \gg f_n$, so that for the quasinuclear branch formula (1) can be rewritten in the form

$$\frac{f_n^2}{f_{n0}^2 - f_n^2} = \frac{f_{e0}^2}{f_T^2}, \quad \text{where} \quad \begin{aligned} f_{e0}^2 &= \gamma_e^2 H(H + H_D) && \text{for } MnCO_3 \\ f_{e0} &= \gamma_e H && \text{for } CsMnF_3, \end{aligned} \qquad (2)$$

where H_D is the Dzyaloshinskiĭ field. We have separated here the parts that depend on the frequency and on the magnetic field. The experimentally observed NMR spectra were therefore reduced in terms of the corresponding coordinates. Figure 1 shows the $MnCO_3$ NMR spectra measured at constant temperature. It is seen that the agreement with the theoretically predicted functional relation of the resonant frequency and the field is satisfactory at all temperatures. From the slopes of the lines in Fig. 1 we can determine f_T^2 and accordingly the average moment of the Mn^{55} nuclei. These data are shown in Fig. 2 as functions of the temperature (triangles). The same figure shows the values of $\langle m\rangle$ recalculated from the plots of $H_{res}(T|_{f_n=\text{const}})$ obtained in the course of cooling of the dissolution cryostat (triangles—f_n = 549 MHz, circles—f_n = 485 MHz). The figure shows also the analogously obtained data for $CsMnF_3$ (squares—f_n = 580 MHz). It is seen that all the results fit satisfactorily one universal curve. This curve differs strongly from the Brilluoin function $B_{5/2}(m_n H_n/kT)$, which describes the polarization of the noninteracting moments

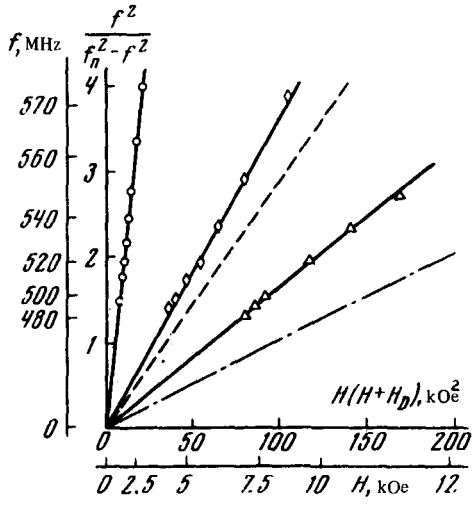

FIG. 1. NMR spectra in $MnCO_3$ at $T = 1.12$°K (○), $T = 0.15$°K (◇), and $T = 0.043$°K (△). Dashed curves—calculation for weakly-interacting moments.

in a hyperfine field (dashed curve in Fig. 2). The result obtained by us are satisfactorily described by the empirical formula (solid curve in Fig. 2)

$$\frac{\langle m \rangle}{m_n} = B_{5/2}\left(\frac{m_n H_n}{kT}\right) - \frac{1}{2}\left[B_{5/2}\left(\frac{m_n H_n}{kT}\right)\right]^{3/2}, \tag{3}$$

where m_n is the magnetic moment of the Mn^{55} nucleus, H_n is the hyperfine field, and k is the Boltzmann constant.

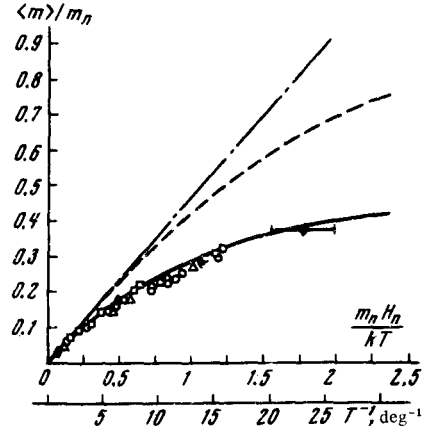

FIG. 2. Average nuclear moment as a function of temperature. Dash-dot—extrapolation of the Curie law.

In principle, this discrepancy can be due to overheating of the samples relative to the helium mixture. One of the causes of the overheating may be the RF pulses used for the measurement. For the pulses used by us ($P \approx 10^{-3}$—10^{-4} W, $t = 1$ μsec) the spin rotation angle due to one pulse is $\theta \approx 10^{-3}$ rad. The corresponding change of the magnetic moment is $\Delta\langle m\rangle_g/\langle m\rangle \approx \theta^2 \approx 10^{-6}$. Consequently the error due to the action of a series of 10—30 pulses, which are needed for the measurement, is negligible. At a pulse repitition frequency ≈ 1 Hz, the average power is $P_{av} \approx 10^{-9}$— 10^{-10} W. The maximum possible overheating of the sample due to the Kapitza jump is therefore $\Delta T = (R/ST^3)P_{av} \approx 10^{-3}$ °K at $T = 0.1$ °K (S is the surface area of the sample and R is the constant of the jump). This estimate was made under the assumption that the entire power delivered to the sample is absorbed. There are no grounds for so large an absorption. Indeed, no change in the NMR signal is produced by increasing the pulse repitition frequency to 50 Hz.

Another possible cause of the overheating may be the slow rate of establishment of thermal equilibrium. The rate of establishment of equilibrium with the helium is determined by the Kapitza thermal-resistance time constant $\tau = RC/T^3$, where C is the heat capacity of the sample. At low temperatures, the heat capacity is determined mainly by the nuclear contribution. To obtain the upper bound of the heat capacity we can extrapolate the data obtained for $MnCO_3$ at $T = 4.2$—1.5 °K, [5] where $C \sim T^{-2}$. At $T = 0.1$ °K we obtain $\tau \approx 1$ sec. Although the rates of the nuclear spin-lattice relaxation in $MnCO_3$ and $CsMnF_3$ at helium temperatures are known, [2,3] the data are not sufficient for a reasonable extrapolation to lower temperatures. However, the low rate of establishment of the thermal equilibrium should manifest itself in a drift of the position of the NMR lines with time, something not observed in the experiment. On the basis of the foregoing we propose that the sample temperature is equal to the helium-mixture temperature, and the effect observed by us is not connected with superheating.

Consequently, the process of polarization of Mn^{55} nuclei in easy-plane antiferromagnets does not reduce to a simple alignment in a hyperfine field.[3)] The indirect Suhl-Nakamura interaction between the nuclear spins of the magnetic moments of magnets leads to the appearance of elementary excitations with spatial dispersion—nuclear spin waves. [1] The problem of the temperature dependence of the average nuclear magnetization has not been solved in this case. It is necessary also to consider the question of the ground state of the nuclear spin system in the case of the Suhl-Nakamura interaction. We hope that an investigation of these processes can explain the results of our experiment.

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

[1)] According to our calculations, for easy-plane antiferromagnets formula (1) is satisfied at all values of the nuclear magnetization, accurate to terms $\approx A\langle m\rangle/H_E$.

[2)] The author is grateful to K. N. Zinov'eva for supplying the calibrated thermometer.

[3)] The temperature dependence of nuclear magnetization of Mn^{55} in ferromagnetic Fe_2MnO_4, obtained in similar fashion, is satisfactorily described by a Brillouin function. [6]

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