Nuclear relaxation of ^{19}F in $RbMnF_3$ at very low temperatures

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We have measured the nuclear spin-lattice relaxation time of ^{19}F, in the antiferromagnet $RbMnF_3$, as a function of field and temperature from 0.1 to 3 K. Since most of the energy of the entire system is contained in the ^{55}Mn nuclear system, it may be viewed as the reservoir. A new relaxation mechanism is suggested whereby ^{19}F Zeeman energy is transferred directly to the ^{55}Mn system. It is found that $T_1(F) \propto \Omega_{10}^2$, where Ω_{10} is the frequency of the field-dependent antiferromagnetic-resonance mode.

I. INTRODUCTION

We report here measurements of the ^{19}F spin-lattice relaxation time T_1 in the antiferromagnet $RbMnF_3$ as a function of the external magnetic field H_0 and temperature in the vicinity of the magnon gap temperature T_{AE}. This work was motivated by the observation of a different relaxation mechanism at low temperatures in $RbMnF_3$ by Hess and Hunt[1] and in $KMnF_3$ by Mahler *et al.*[2]

It was expected that $RbMnF_3$ would provide the best test for the theory of relaxation by antiferromagnetic magnons because its magnon gap temperature is far below the Néel point making a large range where the magnon theory is applicable. It has a simple cubic magnetic structure and is well described by the Heisenberg Hamiltonian.[3] The neutron-diffraction results[3] provide a textbook example. There have been detailed investigations by antiferromagnetic resonance (AFMR)[4] and ultrasonics.[5] Measurements have also been made of the specific heat.[6]

The nuclear-spin-lattice relaxation time T_1 characterizes the rate at which the nuclear-spin energy is transferred to the crystal lattice. In magnetic materials the most probable mechanism for energy transfer is through the hyperfine and dipolar interactions to the magnon system which is presumed in good thermal contact with the "bath." The one-magnon process, in which a nuclear spin relaxes by generating a magnon, is forbidden in antiferromagnets by conservation of energy because of the energy gap ($T_{AE} \sim 0.4$ K in $RbMnF_3$). Thus the most likely processes are those involving two or more magnons.

The most convincing agreement between theory and experiment occurs in the case of MnF_2.[7] Both the temperature and field dependence of the ^{19}F relaxation are explained quantitatively by the two-magnon process and the field dependence of the ^{55}Mn by the three-magnon process below the spin-flop field. Good agreement for the temperature dependence of nuclear-spin-lattice relaxation has been found in other antiferromagnets by Lowe and Whitson[8] using these same mechanisms. For all of these relaxation mechanisms T_1 strongly depends on temperature and is relatively insensitive to the magnetic field. The two-magnon process gives[9] $1/T_1 \propto T^3$ for $T \gg T_{AE}$ and $T^2 c^{-T_{AE}/T}$ for $T \ll T_{AE}$. The exponential decrease has been observed in the case of MnF_2.[10] The three-magnon process depends even more strongly on temperature. In $RbMnF_3$ a strong dependence on T was found above 8 K, but below that the rate varied much more slowly and showed a strong field dependence.[1] This is indicative of a new relaxation mechanism as was pointed out by Mahler *et al.*[2] for the case of $KMnF_3$.

Here we report results of measurements of T_1 between 0.1 and 3 K for fields H_0 between 1 and 5 kOe. In addition we suggest a new relaxation mechanism whereby the ^{19}F nuclei relax directly to the ^{55}Mn nuclear-spin system which contains most of the energy in our temperature range. Finally we show that T_1 within experimental error is proportional to Ω_{10}^2, the square of the AFMR frequency of the field-dependent mode.

II. EXPERIMENTAL

The temperature range was achieved using a ^3He-^4He dilution refrigerator. The sample was placed inside the mixing chamber to assure good thermal contacts with the bath. Carbon and germanium resistance thermometers as well as a heater were attached to the outside of the mixing chamber. To reduce the thermal boundary resistance between the fluid and mixer walls, 260 No. 40 copper wires were attached increasing the contact area to 200 cm^2.

Three methods were used to cover the temperature range. From 1.3 to 3 K a quantity of He gas was admitted to the refrigerator for thermal con-

tact to the ^4He bath whose pressure was controlled. The range from 0.7 to 1.3 K was controlled by evaporating a small amount of the ^3He-^4He mixture in the mixing chamber. Below 0.7 the refrigerator was operated in the dilution mode. These methods were found preferable to using the heater because the large cooling power of the refrigerator necessitated large heating powers and therefore the possibility of large temperature gradients exists.

The carbon resistor was used as the primary thermometer. It was calibrated using a Ge resistor which was calibrated down to 0.26 K using the vapor pressures of ^3He and ^4He. Below this temperature the ^{19}F nuclear magnetization, which obeys Curie's law, was measured by NMR. Measurements taken in the range of the Ge resistor were consistent to ±1%. Curie's law would be expected to hold for temperature greater than $T_0 \sim \mu H_a/k_B$, where H_a is any field tending to align the spins. For our case since the hyperfine field of the Mn^{++} ions cancels at the ^{19}F site and the nuclear dipole interactions are small, the largest field is the applied field. Therefore $T_0 \sim 10^{-3}$ K which is a factor of 10^2 lower than our lowest temperature.

The T_1 measurements were made by saturating the resonance with pulses and monitoring the recovery by cw technique. The exponential recovery signal was stored in a digital memory oscilloscope and could be compared directly to a calibrated exponentially varying voltage derived from an RC circuit. We have found this technique to give more reliable results more quickly than others we have used, as well as permitting recognization of nonexponential recoveries.

III. RESULTS

The $RbMnF_3$ samples are the same two used in earlier experiments and are described in Ref. 1. The data given here are obtained from the 5-mm cube except where noted.

Most of the data were obtained with H_0 in the [111] direction. This is the unique direction in which all three ^{19}F nuclei in a unit cell are magnetically equivalent and a single resonance line results. This serves as a check on crystal orientation and eliminates problems caused by cross relaxation which occurs when two or more resonances are close together. Furthermore, the magnetization direction giving the lowest energy does not abuptly "flop." The angle between M and H_0 varies continuously from 73.3° to 83.8° for H_0 varying between 1.07 and 4.20 kOe.[4]

The relaxation times of ^{19}F from 0.115 to 3 K for five values of the external field in the [111] direction are shown in Fig. 1. The lines will be discussed in Sec. IV. The estimated error is ±5% on each of the points. Below 0.4 K the data show that $T_1 \propto 1/T$ approximately.

The field dependence is shown in Fig. 2 for the temperatures 0.2 and 1.0 K. The lines shown vary as H_0^2 and are fitted to the lowest field points. It is apparent that T_1 varies slightly more rapidly than H_0^2.

To check for the possibility of impurity-dominated relaxation, measurements were made on a second, larger sample of reportedly somewhat higher purity. Because of its cut it was only possible to align it such that H_0 is parallel to a [100] axis. For this orientation the resonance splits into two lines designated by HFL (high-field line) and LFL in Fig. 3. The splitting is proportional to the net magnetization and hence proportional to H_0. At 4.2 kOe the lines are well separated. For the LFL the two samples yield indistinguishable data, but those for the HFL are about 20% longer for the large sample. We believe this difference is due to the presence of domains which have been shown to exist[11] and, to some extent, remain after repeated efforts to remove them.

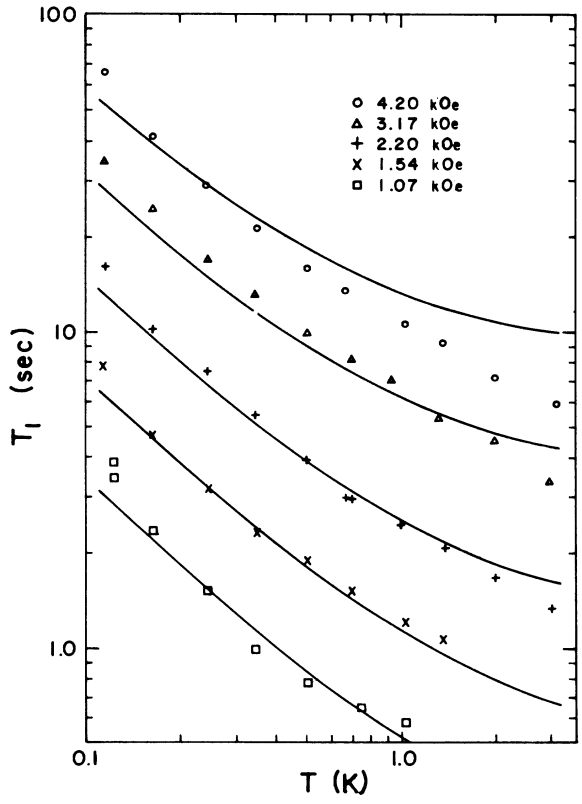

FIG. 1. Temperature and field dependence for the spin-lattice relaxation time at ^{19}F in $RbMnF_3$. The field is in the [111] direction. The lines are obtained from the equation $T_1 \propto \Omega_{10}^2 H_0^2$ normalized at $T = 0.5$ K, $H_0 = 2.2$ kOe.

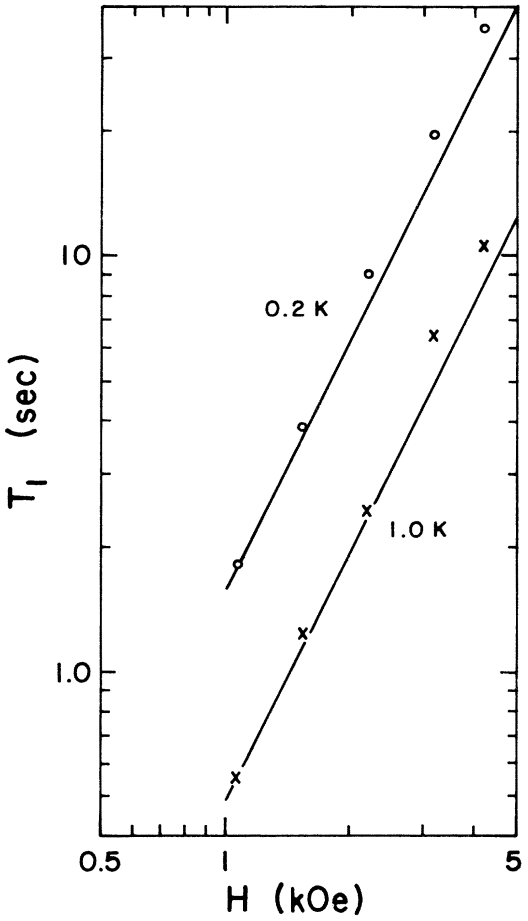

FIG. 2. Field dependence at $T=0.2$ and 1.0 K as obtained from Fig. 1. The lines are proportional to H_0^2, and are fitted at the lowest field points.

IV. DISCUSSION

Both the measured field and temperature dependence are far from what is expected if the relaxation mechanism were associated with magnon processes. Near $T_{AE} \sim 0.4$ K the number of magnons begin to decrease exponentially which should have a similar effect on $1/T_1$, but no such effect is observed. Also the expected magnon processes are for the most part independent of H_0. Other mechanisms such as relaxation to combined magnon-phonon modes or a one-magnon process with damping also meet with some of the same difficulties and do not give a reasonable magnitude for T_1.

The mechanism we suggest here is that the ^{19}F nuclei relax directly to ^{55}Mn nuclear-spin system. The ^{55}Mn nuclei account for most of the specific heat of the crystal below 2 K,[6] and contain about 7×10^3 more energy than the ^{19}F system in our highest field, and, therefore, can be considered as the bath. Furthermore, since the electronic magnetization is "flopped," that is, perpendicular to the applied field, for practical purposes, the ^{55}Mn nuclei are also quantized perpendicular to H_0 and thus to the ^{19}F spins. Thus changes in m_I of the ^{55}Mn nuclei produce changes in the transverse field at the ^{19}F site by means of the dipolar interaction.

The order of magnitude of the relaxation time can be estimated by assuming only the two nearest-neighbor Mn nuclei independently contributed by the equation[12]

$$\frac{1}{T_1} = 2\gamma_F^2 \langle H_x^2 \rangle_{av} \frac{\eta^{-1}}{1+\omega_0^2\eta^{-2}}, \tag{1}$$

where the mean-square dipolar field at the ^{19}F site due to ^{55}Mn nucleus is $\langle H_x^2\rangle_{av} = \frac{1}{3}I(I+1)(2\gamma_{Mn}h/r^3)^2 = (2.5\ \text{Oe})^2$, γ_F is the ^{19}F gyromagnetic ratio, $\omega_0 = \gamma_F H_0$, and η is the rate the ith Mn nucleus changes m_I values defined by the equation

$$\langle I_i^z(t) I_i^z(0)\rangle = \tfrac{1}{3}I(I+1)e^{-\eta t}. \tag{2}$$

One should not view η as simply the reciprocal of the spin-lattice relaxation time (or in our case the spin-^{3}He-^{4}He mixture thermal equilibrium time) which would be its limiting value. An individual spin may flip and exchange its energy with a neighboring spin at a much greater rate by broadening mechanisms—dipole-dipole or the Suhl-Nakamura interaction.[13,14] Therefore, as one views a pair of dipoles mutually flipping at a rate determined by the dipolar interaction, we associate at this point η with the Mn NMR linewidth $\Delta\nu$ ($\eta = 2\pi\Delta\nu$, where $\Delta\nu$ is the half width at half maximum).

The ^{55}Mn linewidth has been measured, and at high fields where inhomogeneities in the electronic

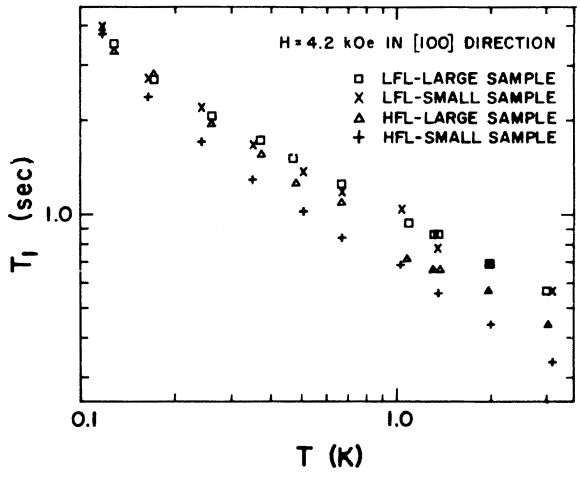

FIG. 3. Relaxation time for two samples as a function of temperature for $H_0 = 4.2$ kOe in the [100] direction.

system have little effect, Freiser *et al.*[15] give $\Delta\nu \sim 0.28$ MHz ($\eta = 1.76 \times 10^6$ sec^{-1}) at 4.2 K. Weber and Seavey[16] confirm this and give values that show $\Delta\nu$ has decreased almost a factor of 2 at 1.79 K. Noting that $\omega_0^2\eta^{-2} \gg 1$ for all our fields, Eq. (1) becomes simply

$$1/T_1 = 2(\langle H_x^2\rangle_{av}/H_0^2)\eta \qquad (3)$$

and at 4.2 K, $H_0 = 3.2$ kOe, Eq. (2) gives $T_1 = 0.5$ sec. The experimental value under these conditions is 3 sec. We further note the measured increase in T_1 to 5 sec at 1.8 K is accounted for by the decrease in $\Delta\nu$, and that the field dependence is essentially correct if η were independent of H_0.

The close agreement found here is encouraging since no other mechanism that we know of gives the correct magnitude. Unfortunately, in our field range Weber and Seavey's data[16] is strongly field dependent, $\Delta\nu$ being proportional to $1/H_0^3$. This effect is interpreted in terms of inhomogeneities in the electronic system. The linewidth is proportional to the AFMR linewidth which is about 150 Oe for their sample and about 100 Oe for a sample from the same source as ours.[4] However, this field-dependent broadening may not be a factor in determining the ^{19}F relaxation rate. The rate depends primarily on the difference between the ^{55}Mn auto- and pair-correlation functions, whereas the ^{55}Mn linewidth depends on all possible correlation functions.

That we have overaccounted for the ^{19}F relaxation rate is reasonable in that we have neglected correlations between neighboring ^{55}Mn nuclei. In fact, there is a high degree of correlation. Hinderks and Richards[17] have observed ^{55}Mn nuclear spin waves in RbMnF$_3$ at 1.15 K by the parallel pumping technique. They have furthermore inferred a k-dependent lifetime η_k. For $k \to 0$, the relaxation rate η_0, which they associated with the NMR linewidth, is 1.4×10^6 sec^{-1} in reasonable agreement with the values used above.

In order to account for the temperature dependence of T_1, the ^{55}Mn nuclear-spin correlation functions must be known. Richards[18] has calculated η_k and its temperature dependence for RbMnF$_3$. His results, although not directly applicable to our problem, suggest a function of the form $\eta(\Omega_{10}, T)$. We have found that using $\eta = 1.33 \times 10^{27}\ \Omega_{10}^{-2}$ in Eq. (3) describes the observed temperature dependence very well, and also accounts for the extra field dependence. The solid lines in Fig. 1 were calculated using values of Ω_{10} for H_0 in the [111] direction obtained from Ref. 4, and were normalized at $H_0 = 2.2$ kOe and $T = 0.5$ K. The departure of the lines from the data at high temperatures is due to the onset of another relaxation mechanism[1] at about 5 K. The excellent agreement is compelling evidence that the relaxation mechanism is an intrinsic one and further theoretical work is indicated.

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