the interpretation of the x-ray data, and assume that at high concentrations (≥ 65 at.% Mn) the (B) site becomes at least partially occupied. The linewidth data could be explained by assuming that the (A)-site magnetic interactions (probably AOA superexchange) are greater than (B)-site interactions (possibly AOB or BOB).

Our results corroborate the susceptibility measurements of Wickham *et al.*[23] on $Mn_{2x}Mg_{2-2x}SnO_4$ spinels. They found that replacing (B)-site Mn^{2+} with Mg^{2+} increased the magnetic moment while we observe a leveling off in ω_E/γ by increasing Mn^{2+} on (B) sites.

The present investigation extends the work of Anderson and Weiss[7] since we now consider a more realistic line shape as well as inherent spin-packet linewidths. The method complements the ESR pair-interaction work of Coles *et al.*[24] and Owen[25] by vastly extending the range of Mn^{2+} concentration.

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[1]B. Bleaney and D. J. E. Ingram, Proc. Roy. Soc. (London) A205, 336 (1951).

[2]W. D. Hershberger and H. N. Leifer, Phys. Rev. 88, 714 (1952).

[3]J. S. Van Wieringen, Discussions Faraday Soc. 19, 118 (1955).

[4]R. Stahl-Brada and W. Low, Phys. Rev. 113, 775 (1959).

[5]F. Waldner, Helv. Phys. Acta 35, 542 (1962).

[6]P. W. Anderson, J. Phys. Soc. Japan 9, 316 (1954).

[7]P. W. Anderson and P. R. Weiss, Rev. Mod. Phys. 25, 269 (1953).

[8]J. D. Dunitz and L. E. Orgel, J. Phys. Chem. Solids 3, 20 (1957); 3, 318 (1957).

[9]D. S. McClure, J. Phys. Chem. Solids 3, 311 (1957).

[10]J. H. Van Vleck, J. Chem. Phys. 3, 803 (1935).

[11]A. Navrotsky and O. J. Kleppa, J. Inorg. Nucl. Chem. 29, 2701 (1967).

[12]S. Greenwald, S. J. Pickart, and F. H. Grannis, J. Chem. Phys. 22, 1597 (1954).

[13]E. O. Schultz-Dubois, Bell System Tech. J. 38, 271 (1959).

[14]R. H. Sands, Phys. Rev. 99, 1222 (1955).

[15]J. D. Swalen and H. M. Gladney, IBM J. Res. Develop. 8, 515 (1964).

[16]L. E. Mohrmann, Jr., B. B. Garrett, and W. B. Lewis, J. Chem. Phys. 52, 535 (1970).

[17]E. Simanek and K. A. Mueller, Chem. Phys. Letters 4, 482 (1970).

[18]R. D. Dowsing, J. Mag. Res. 2, 42 (1970).

[19]F. Walder (private communication).

[20]J. H. Van Vleck, Phys. Rev. 74, 1168 (1948).

[21]C. P. Poole, Jr., *Experimental Techniques of Electron Spin Resonance* (Wiley, New York, 1967).

[22]H. A. Farach, E. F. Strother, and C. P. Poole, Jr., J. Phys. Chem. Solids 31, 1491 (1970).

[23]D. G. Wickham, N. Menyuk, and K. Dwight, J. Phys. Chem. Solids 20, 316 (1961).

[24]B. A. Coles, J. W. Orton, and J. Owen, Phys. Rev. Letters 4, 116 (1960).

[25]J. Owen, J. Appl. Phys. 32, 213S (1961).

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Nuclear-Resonance Study of F^{19} in $RbMnF_3$†

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A nuclear-magnetic-resonance study is made of the F^{19} nucleus in the nearly isotropic antiferromagnet $RbMnF_3$ from 1.5 to 300 K. In the paramagnetic region the results are in satisfactory agreement with Moriya's theory applied to this material. In the antiferromagnetic region the spin-lattice relaxation time can be empirically described by an Arrhenius equation over four orders of magnitude. Below 30 K the spin-spin relaxation time is anomalously long—three times that calculated using dipolar interactions alone.

I. INTRODUCTION

We present fairly detailed measurements of the F^{19} nuclear magnetic resonance in the nearly isotropic antiferromagnet $RbMnF_3$ from 1.5 to 300 K. Where possible we have used pulse methods to measure the spin-lattice relaxation time T_1 and the spin-spin relaxation time T_2. These techniques discriminate against inhomogeneous broadening and thus give more reliable data than steady-state

linewidth studies. These measurements are of interest because they give information on the dynamics of the Mn^{++} ionic spin fluctuations.

$RbMnF_3$ has the perovskite structure—the Mn^{++} ions are at the corners of a cube with the Rb^+ ion at its center and the F^- ions are centered on the cube edges.

Moriya[1] has given a theory of nuclear relaxation in magnetic materials where the relaxation is caused by the modulation of the hyperfine interaction by the exchange interaction. The central theoretical problem lies in the evaluation of the various ionic spin-correlation functions. These functions have been approximated at infinite temperature (the spins are assumed to be independent) and at low temperatures (the spin-wave approximation). Walker[2] has applied the theory to our case, the F^{19} resonance in $RbMnF_3$, using the high-temperature approximation.

In a later paper Moriya[3] uses the molecular field approximation to evaluate the correlation functions in the paramagnetic region. To our knowledge there have been no attempts to verify this theory except near the transition temperature,[4] where the molecular field approximation is known to fail. We apply this theory to our case and find acceptable agreement with our experimental results both in magnitude and temperature dependence. The molecular field approximation is adequate in our case even at T_N because the hyperfine interaction with the two nearest-neighbor Mn^{++} ions vanishes for the antiferromagnetic mode.

In the antiferromagnetic region we find T_1 to be given empirically by a simple equation and T_2 to be anomalously long. From the Néel temperature ($T_N = 82.9$ K) to about 8 K, we find T_1 can be represented by an Arrhenius equation with an activation energy of about $k_B T_N$. Furthermore, $T_2 = T_1$ down to about 35 K, where T_2 attains a temperature-independent value about three times longer than that calculated from dipolar interactions alone.

Moriya's theory is applied to our case in Sec. II. A brief description of the sample and experiment is given in Sec. III and the experimental results are presented and discussed in Sec. IV.

II. THEORY–PARAMAGNETIC REGION

The relaxation of the F^{19} nuclei is assumed to be caused by the hyperfine interaction with the Mn^{++} ions. The interaction between the pth F nuclear spin and the jth Mn^{++} ionic spin is $\vec{I}_p \cdot \overleftrightarrow{A}_{pj} \cdot \vec{S}_j$, where $\overleftrightarrow{A}_{pj}$ includes both the hyperfine and dipolar interactions. Moriya[3] gives the expression for T_1 as

$$\frac{1}{T_1} = \frac{1}{2\hbar^2} \int_{-\infty}^{\infty} dt \cos\omega_0 t \sum_{j,j'} \sum_{\nu,\nu'} [(A_{pj}^{x\nu}) + i(A_{pj}^{y\nu})] \times [(A_{pj'}^{x\nu'}) - i(A_{pj'}^{y\nu'})] \langle \{ \delta S_{j\nu}(t)\, \delta S_{j'\nu'}(0) \} \rangle , \quad (1)$$

where ω_0 is the NMR frequency, ν, ν' represent x, y, z, $\delta\vec{S}_j = \vec{S}_j - \langle S_j \rangle$, $\langle\ \rangle$ denotes a thermal average of the spin functions and $\{AB\} = \frac{1}{2}(AB + BA)$.

The principal difficulty is the evaluation of the correlation functions. Walker,[2] using the Heisenberg Hamiltonian $\sum_{j<j'} J_{jj'} \vec{S}_j \cdot \vec{S}_{j'}$ to describe the spin system, makes an infinite temperature approximation for T_2 by ignoring correlations between different spins and assuming the autocorrelation function decays like $e^{-\omega_e^2 t^2/2}$, where $\hbar^2\omega_e^2 = \frac{2}{3} S(S+1) \times \sum_j J_{jj'}^2$. The result is a Lorentzian line shape with

$$\frac{1}{T_{2\infty}} = \left(\frac{\pi}{2}\right)^{1/2} \frac{S(S+1)}{3\hbar^2\omega_e} \sum_{j,\nu} [(A_{pj}^{z\nu})^2 + \tfrac{1}{2}(A_{pj}^{x\nu})^2 + \tfrac{1}{2}(A_{pj}^{y\nu})^2] . \quad (2)$$

If the F^{19}-Mn^{++} hyperfine interaction were isotropic, $T_1 = T_2$.[5] However, there is some anisotropy due to the Mn^{++} dipole moment.[6] For the purpose of calculating the temperature dependence of the linewidth we shall neglect the anisotropy and assume that T_1 and T_2 are equal.

Silbernagel *et al.*[7] have pointed out the importance of pair correlations at finite temperatures. Carrying out a calculation for $RbMnF_3$ similar to theirs for the F^{19} resonance in MnF_2, we find

$$\frac{T_{1\infty}}{T_1} \simeq \frac{T_{2\infty}}{T_2} \simeq 1 - \frac{1}{4}\frac{T_N}{T} . \quad (3)$$

The temperature dependence comes from the high-temperature expansion of the pair-correlation function. Our temperature range includes T_N and a better approximation is needed.

Moriya puts the Fourier transform of the correlation functions in terms of $\chi(\vec{k})$, the wavelength-dependent susceptibilities, and a relaxation function characterized by an exponential decay with a rate $\Gamma_{\vec{k}}$. Equation (1) applied to our case, assuming an isotropic nearest-neighbor hyperfine interaction of strength A, becomes

$$\frac{1}{T_1} = \frac{4A^2 k_B T}{Ng^2\mu_B^2\hbar^2} \sum \frac{\chi(\vec{k})}{\Gamma_{\vec{k}}} (1 + \cos k_x a) . \quad (4)$$

The functions $\chi(\vec{k})$ and $\Gamma_{\vec{k}}$ and the notation are given in Ref. 3. The applied field is in the [001] direction and we consider the F^{19} nuclei whose two nearest-neighbor Mn^{++} ions are in the [100] and [$\bar{1}$00] directions. The unusual feature in this equation is the factor $(1 + \cos k_x a)$, which prevents $1/T_1$ from getting large near the antiferromagnetic mode $\vec{K}_0 = (\pi/a, \pi/a, \pi/a)$ and T_N. This can be seen since for small $\vec{k}$ about $\vec{K}_0$,

$$\chi(\vec{K}_0 + \vec{k})/\Gamma_{\vec{K}_0+\vec{k}} = C/(\epsilon + \gamma k^2)^2 ,$$

from Ref. 3. Here $\epsilon = (T - T_N)/T_N$ and C and γ are constants. Converting the sum to an integral, the integrand becomes constant as $\epsilon \to 0$ and the

F^{19} linewidth is expected to remain finite in that limit.

In evaluating Eq. (4) we find no one region of k space dominates and therefore the integral cannot be approximated. The sum in Eq. (4), however, can be put in terms of the function

$$I_\epsilon = N^{-1}\sum_k [1+\epsilon - J(\vec{k})/J(\vec{K}_0)]^{-1}$$

and its derivative. Here we have

$$J(\vec{q}) = 2J(\cos q_x a + \cos q_y a + \cos q_z a)$$

for the simple-cubic magnetic lattice. We then find

$$\frac{T_{1\infty}}{T_1} = \frac{2(I_0+I_\epsilon)(2+\epsilon)^{-2} + \epsilon(\partial I_\epsilon/\partial\epsilon)(2+\epsilon)^{-1}}{6[(1+\epsilon)I_\epsilon - 1](2I_0 - 1)} \quad . \quad (5)$$

The quantities I_ϵ and $\partial I_\epsilon/\partial\epsilon$ have been computed by Mannari and Kawabata.[8]

III. EXPERIMENTAL

The measurements were made on a 5-mm cube supplied by the MIT Center for Materials Science. A mass spectrographic analysis of crystals grown from the same starting materials showed Fe and Co contents of 40 and 50 ppm (parts per million), respectively, with an estimated accuracy of 4 ppm. Some measurements were repeated with a reportedly somewhat purer sample obtained from the Cornell Crystal Growing Facility.

The anisotropy of the hyperfine interaction splits the F^{19} resonance generally into three lines. Our measurements are taken with H_0 in the [001] direction for which two of the three lines coincide. The data given are for the stronger of the two and correspond to those nuclei whose two nearest-neighbor Mn^{++} ions lie on a line perpendicular to the applied field. The merging of the three lines for the field in the [111] direction served as a rather sensitive ($\sim 1^\circ$) check on the alignment of the crystal.

The pulse spectrometer is conventional. A 90° pulse can be attained in 3.5 μsec and the amplifier recovers in another 1.5 μsec. Since the signal following a 90° pulse can be observed when the sample is below T_N, the Carr-Purcell[9] pulse sequences were used for the T_1 and T_2 measurements. In the paramagnetic temperature region the resonance is too broad for our pulse apparatus and linewidths were measured by standard steady-state resonance techniques.

The sample and thermometer are enclosed in a Cu can which is contained in a double Dewar system. The sample is thermally and physically connected to the lid of the can by means of a Cu support. "Double-stick tape" is used to fasten the sample to the support. The lid has several small holes drilled into it to allow gas in the inner Dewar to provide an additional thermal connection between the sample and can. A heater wire is wrapped uniformly around the can.

Several different methods of controlling temperature were employed. For $T \leq 4.2$ K the can was immersed in liquid He. Stable temperatures between 4.2 and 20 K were obtained by balancing a heat input against the heat lost to some liquid remaining in the tail of the Dewar. Once the liquid disappeared, the system warmed sufficiently slowly so measurements could be made on the run. Temperatures between 45 and 77 K were attained by pumping on liquid and solid N_2 contained in the He Dewar. Above 77 K the heat leak to the outer N_2 Dewar was balanced with the heater.

Two thermometers were used—a commercially obtained, calibrated Ge resistance thermometer (Cryocal, Inc.) for $T < 100$ K and a Cu-wire thermometer[10] for $T > 50$ K. Both were fastened securely to the lid of the can. The calibration was verified for the Ge thermometer and was made for the Cu thermometer using the vapor pressures of He, N_2, and O_2. For the Cu wire one further point was obtained at room temperature. The Ge resistor has a small magnetic field dependence which was determined at several fixed temperatures where the vapor pressure of the coolant could easily be measured. Intermediate corrections were interpolated. We estimate the maximum uncertainty in the temperature to be ± 0.1 below 4 K and from 56 to 93 K where vapor pressure measurements were made, ± 0.5 K in the range 4 to 45 K because of difficulties in attaining stable or uniform temperatures, and ± 0.2 K from 45 to 56 K where steady temperatures without heat were attained but no independent calibration was made. Above 93 K we estimate ± 1 K, but no checks were made since errors of even several degrees would not affect our conclusions.

The Néel temperature was obtained for our sample by observing the appearance and disappearance of the paramagnetic resonance at a frequency of 8 MHz. The value obtained is 82.9 ± 0.1 K and is in good agreement with other determinations, the most recent of which range from 82.93 to 82.99 ± 0.01 K for three crystals.[11]

IV. RESULTS AND DISCUSSION

A. Paramagnetic Region

The results of the steady-state linewidth measurements are shown as circles in Fig. 1. The linewidth ΔH is measured between the extrema of the derivative of the absorption. The frequency used here, 8.9 MHz, is a compromise between keeping separate the two resonances from inequivalent sites and minimizing the broadening due to inhomogeneous demagnetizing effects. The broadening arises because the sample has a cubic shape. The quantity $4\pi M$, where M is the magnetization,

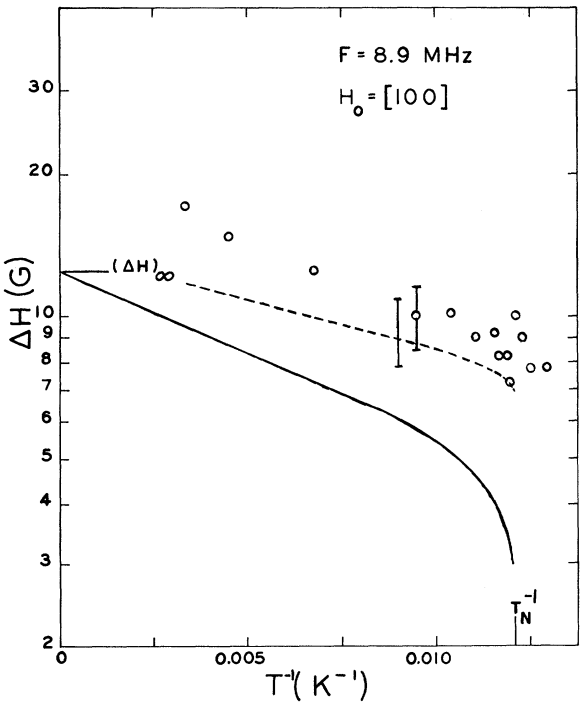

FIG. 1. Measured linewidth ΔH of the stronger of the two lines (circles) as a function of inverse temperature in the paramagnetic region. The solid line represents the theory [Eq. (5)] and the dashed line represents the corrected theoretical linewidth. The correction is made by convoluting the theoretical Lorentzian line shape with a Gaussian function whose width is calculated from dipolar interactions.

ranges from 7 to 10 G in our field of 2.2 kG, and is of the same magnitude as the linewidths. However, the observed linewidths were found to be field independent until the field was increased above 3 kG. Unfortunately, the spin-echo method could not be used here because T_2 (~ 5 μsec) is about the same as our pulse widths.

At room temperature there is good agreement between our results and those of others. Our value of ΔH is 17 ± 2 G, and under the same conditions Walker and Stevenson[6] found 17 ± 1 G and Melcher and Bolef[12] found 17 ± 2 G. At 90 K we found 9 ± 1 G and Walker and Stevenson found 10.5 G, but Melcher and Bolef reported 13.5 ± 1 G and thus have some extraneous broadening. Extrapolating our data to infinite temperature, we obtain 22 ± 2 G, in excellent agreement with the value 22.7 ± 1 G obtained by Gulley *et al.*[13]

The theory of Sec. II is also shown in Fig. 1 as the solid line. Here we have used the fact that $\Delta H = 2/\sqrt{3}\,\gamma T_2$ for the Lorentzian line shape. Using the value for the exchange integral J, 6.8 ± 0.6 K,[14] and the values of $A_{pj}^{\nu\nu'}$ from Ref. 6, $T_{2\infty}$ is found from Eq. (2) to be 3.7 ± 0.6 μsec ($\Delta H = 12 \pm 2$ G). The temperature dependence is calculated from Eq. (5) assuming $T_1 = T_2$, that is, neglecting the anisotropic part of the hyperfine interaction.

However, in order to compare our results with the theory of Sec. II, we must take into account that part of the linewidth is due to the nuclear-dipole interactions. We can calculate the contribution to the second moment, M_2, using the Van Vleck formula, but then we must assume some line shape in order to get the total theoretical linewidth.

The dipolar contribution arises from interactions with (a) the two kinds of F^{19} nuclei on magnetically equivalent sites, (b) the F^{19} nuclei on inequivalent sites, (c) the Rb^{85} and Rb^{87} nuclei, and (d) the Mn^{55} nuclei. We obtain 5.4×10^8 and 14.9×10^8 rad^2/sec^2 for (a) types, and, treating (b) types as "unlike spins" because they give rise to a distinct resonance, we obtain 1.9×10^8 rad^2/sec^2. Type-(c) spins contribute 0.5×10^8 rad^2/sec^2 and type-(d) spins 20.6×10^8 rad^2/sec^2. The total contribution to the rms width comes to 2.6 G.

The measured line is a composite of a Lorentzian line whose width is due to the Mn^{++} fluctuations broadened by the nuclear-dipolar interaction whose shape function is unknown but is probably nearly Gaussian. A computer was used to convolute Lorentzian lines of various widths with a Gaussian line of rms width 2.6 G and to print out the resultant line shape. From these line shapes values of the total theoretical linewidth were obtained and are shown by the dashed line in Fig. 1. Within the rather large uncertainties (16% for the theory, taking into account only errors in J and $A_{pj}^{\nu\nu'}$, and 15% for the experiment) the agreement is satisfactory. There are no adjustable parameters. Near the critical temperature we find a narrowing of the line instead of the usual broadening.[15] As we mentioned earlier, this is because the F^{19} nuclei do not interact with the antiferromagnetic mode.

Unfortunately, the uncertainty in our data does not permit a good comparison with the high-temperature expansion given by Eq. (3). However, the fact that experimentally T_2 (or T_1) increases with decreasing temperature is in qualitative agreement.

B. Antiferromagnetic Region

The relaxation times T_1 and T_2, measured by pulse techniques, are given in Fig. 2 as a function of temperature. T_1 was measured at the three frequencies shown. The T_2 spin-echo data were obtained at 39 MHz. Not shown is the steady-state line, measured at 13.5 MHz and found to be nearly Lorentzian with a width of 5.5 ± 0.5 G and independent of temperature. This width corresponds to a T_2 value of 8.4 μsec.

1. Spin-Spin Relaxation

Below 30 K the decay of the envelope of the

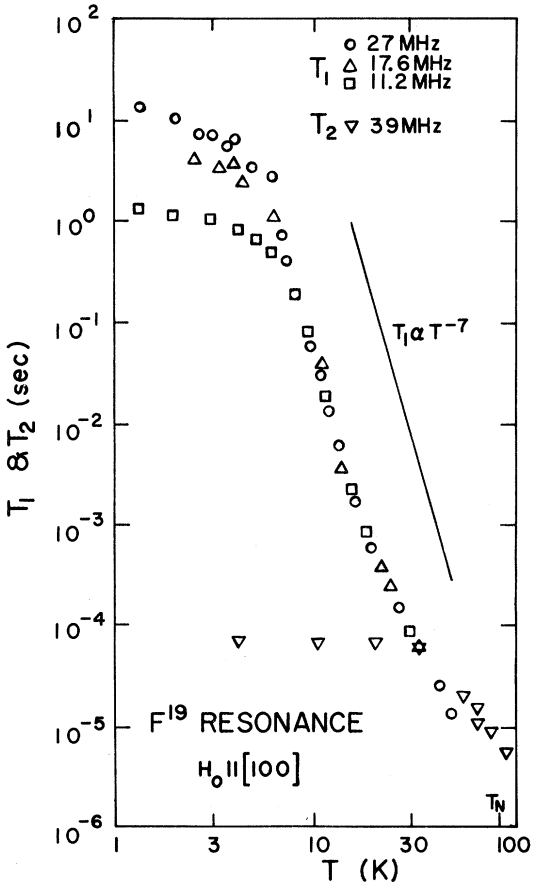

FIG. 2. Relaxation times as a function of temperature in the antiferromagnetic region.

echoes $F(t)$ was found to be independent of temperature and to have a Gaussian shape within experimental error. For a Gaussian decay, we have

$$F(t) \propto \exp\left(-\frac{\pi}{4}\frac{t^2}{T_2^2}\right) = \exp(-M_2 t^2/2) .$$

The value of T_2 thus obtained is 79 ± 8 μsec, which corresponds to an rms width of 0.65 G. The width, being significantly narrower than the steady-state linewidth, normally indicates the presence of inhomogeneous broadening. However, the width is even narrower than that calculated by the dipolar interactions.

The dipolar width is calculated as in Sec. IV A with the following exception: The Mn^{++} moments are aligned perpendicular to the applied field and hence the Mn nuclei are quantized in a direction perpendicular to that of the F^{19} nuclei. Consequently, the component of the Mn nuclear-dipole field along the applied field vanishes at the nearest-neighbor F^{19} sites and is negligible at other sites. Adding the remaining contributions gives a total of 22.7×10^8 rad^2/sec^2, which corresponds to an rms width of 1.89 G. This is nearly a factor of 3 greater than the experimental value.

This unusual circumstance of the calculated T_2 value being shorter than the measured one has been noted before.[16,17] It has been shown that crystal imperfections can disrupt part of the dipolar interaction producing the effect that "like" spins tend to behave like "unlike" spins. Treating all the spins as unlike, however, still makes the calculated width too large by a factor of 2. Thus we are forced to look into other mechanisms for a source of the narrowing. Hone *et al.*[16] have shown that if a Suhl-Nakamura interaction[18] exists and its magnitude is comparable to the dipolar interaction, there can result a substantially reduced M_2 due to destructive interference. This, however, is an open question.

Above 30 K the echo decays become exponential and lifetime broadened ($T_1 = T_2$). Near T_N, T_2 becomes comparable to the rf pulse widths, thereby reducing the reliability of the data.

2. *Spin-Lattice Relaxation*

The break in the behavior of T_1 at 6 K shown in Fig. 2 indicates that there are two relaxation mechanisms present. The low-temperature data are very similar to those found by Mahler *et al.*[19] for the F^{19} resonance in $KMnF_3$. It is characterized by T^{-1} temperature dependence within experimental error. In addition, we find that T_1 becomes proportional to H^2 at our lowest temperature (1.5 K). To our knowledge the relaxation mechanism is not understood.

Between 8 and 16 K the temperature dependence of T_1 can be represented by a T^{-7} behavior within experimental error. This is the same as that found in $KMnF_3$ by Mahler *et al.*[19] for their "impure" sample. They found, however, a T^{-5} dependence, characteristic of a three-magnon process, for a nominally "pure" (30-ppm) sample. This would indicate that our results are due to some sort of crystalline imperfections. We acquired another sample, reportedly somewhat purer, and remeasured T_1 from 1.5 to 20 K with results identical to the first sample. Thus either the impurity level must be about the same in the two samples from different sources, or the effect of the impurity must be negligible.

It is worth looking at the data in another way. The gradual curvature of the data shown in Fig. 2 suggests that the usual power-law representation is not appropriate. In Fig. 3 we make a semilog plot of T_1 vs T^{-1} and find an interesting result. The relaxation rate follows an Arrhenius equation over four orders of magnitude with an activation energy roughly equal to $k_B T_N$. Furthermore, the infinite temperature value of T_1 is the same as that found earlier in the paramagnetic region. The equation of the dotted line in Fig. 3 is

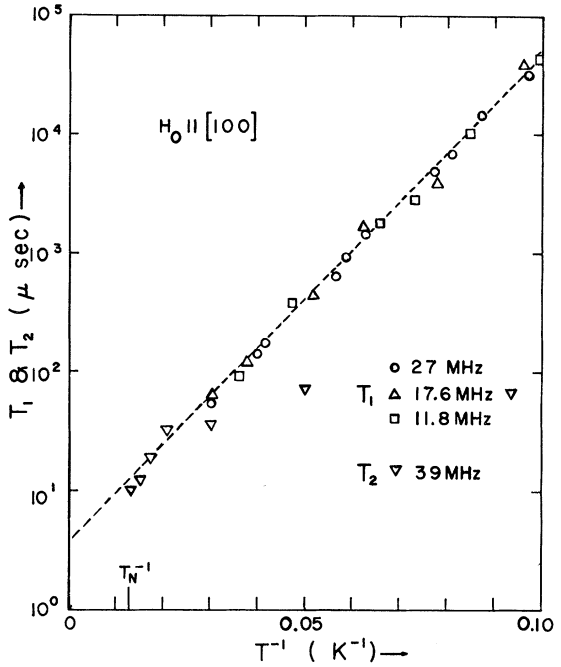

FIG. 3. Relaxation times plotted against inverse temperature. The dotted line represents Eq. (6).

$$1/T_1 = \tfrac{1}{3.7}\, e^{-1.13 T_N/T}\ \mu\text{sec}^{-1}\ . \qquad (6)$$

The above result cannot be explained simply on the basis of a dipolar coupling to magnetic impurity. For this we would expect[20]

$$1/T_1 \propto \tau/(1+\omega_0^2\tau^2)\ ,$$

where τ is the impurity longitudinal relaxation time and ω_0 is the F^{19} resonant frequency. If $\omega_0\tau \ll 1$ we would have the unlikely result that τ gets shorter as the temperature is lowered and if $\omega_0\tau \gg 1$ we would get a frequency dependence contrary to our results. Furthermore, the concentration of impurities would have to be much higher than we would expect on the basis of the analysis and of the consistency of our measured value of T_N with other values.

Because of the symmetry of the crystal the interaction of the F^{19} nuclei with the Mn^{++} spins vanishes as the spins become aligned and thus T_1 becomes longer at lower temperatures. The relaxation rate therefore depends on the breaking up of pair correlations, and Eq. (6) implies a barrier to this whose height is about equal to $k_B T_N$.

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[1]T. Moriya, Progr. Theoret. Phys. (Kyoto) 16, 23 (1956); 16, 641 (1956).

[2]M. B. Walker, Proc. Phys. Soc. (London) 87, 45 (1966).

[3]T. Moriya, Progr. Theoret. Phys. (Kyoto) 28, 371 (1962).

[4]P. Heller, in *Critical Phenomena*, Natl. Bur. Std. (U. S.) Misc. Publ. No. 273, edited by M. S. Green and I. V. Sengers (U. S. GPO, Washington, D. C., 1966).

[5]V. Jaccarino, in *Magnetism*, edited by G. Rado and H. Suhl (Academic, New York, 1965), Vol. IIA, Chap. 5.

[6]M. B. Walker and R. W. H. Stevenson, Proc. Phys. Soc. (London) 87, 35 (1966).

[7]B. G. Silbernagel, V. Jaccarino, P. Pincus, and J. H. Wernick, Phys. Rev. Letters 20, 1091 (1968).

[8]I. Mannari and C. Kawabata, Research Notes of the Department of Physics, Okayama University, No. 15, 1964 (unpublished).

[9]H. Y. Carr and E. M. Purcell, Phys. Rev. 94, 630 (1954).

[10]T. M. Dauphinee and H. Preston-Thomas, Rev. Sci. Instr. 25, 884 (1954).

[11]T. J. Moran and B. Luthi, Phys. Rev. B 4, 122 (1971).

[12]R. L. Melcher and D. I. Bolef, Phys. Rev. 184, 556 (1969).

[13]J. E. Gulley, D. Hone, D. J. Scalapino, and G. G. Silbernagel, Phys. Rev. B 1, 1020 (1970).

[14]L. G. Windsor and R. W. Stevenson, Proc. Phys. Soc. (London) 87, 501 (1966).

[15]Albert M. Gottlieb and Peter Heller, Phys. Rev. B 3, 3615 (1971).

[16]D. Hone, V. Jaccarino, Tin Ngwe, and P. Pincus, Phys. Rev. 186, 291 (1969).

[17]Peter Richards, Phys. Rev. 173, 581 (1968).

[18]H. Suhl, Phys. Rev. 109, 606 (1958); T. Nakamura, Progr. Theoret. Phys. (Kyoto) 20, 542 (1958).

[19]P. J. Mahler, A. C. Daniel, and P. T. Parrish, Phys. Rev. Letters 19, 85 (1967).

[20]A. Abragam, *The Principles of Nuclear Magnetism* (Oxford U. P., London, 1961), p. 380