Nuclear-spin lattice relaxation and magnetic-ion spin fluctuations in Heisenberg antiferromagnets below T_N

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The results of measurements on the magnetic field and temperature dependences of the ^{19}F nuclear-spin lattice relaxation time T_1 in $KNiF_3$ for $T \lesssim 0.04T_N$ are reported. It is concluded that a relaxation mechanism that had been previously proposed to interpret the low-temperature field dependence of T_1 in $RbMnF_3$ does not explain our experimental results in $KNiF_3$. Some similarities in the behavior of both systems suggest that a common mechanism may be responsible for spin-lattice relaxation in either case. We discuss the possibility that this mechanism may involve a diffusive mode below T_N with a central peak in the relevant magnetic-ion spin correlation function.

I. INTRODUCTION

Nuclear-spin lattice relaxation (NSLR) has proven to be a powerful tool for probing the nature of the fluctuations in the local magnetic field experienced by nuclei due to various excitations present in solids. In antiferromagnets NSLR is often achieved via the transferred hyperfine coupling. This interaction allows the scattering of magnons to be accompanied by a nuclear spin flip. A variety of NSLR processes involving magnons have been proposed and identified experimentally.[1,2]

$RbMnF_3$ and $KNiF_3$ are model antiferromagnets whose magnetic properties can be described in some cases to a good approximation by a pure Heisenberg model. The ratios of the anisotropy field to the exchange field are extremely small in these systems. For example, for $RbMnF_3$ $H_A/H_E \sim 5\times10^{-6}$ while for $KNiF_3$ $H_A/H_E \sim 7\times10^{-5}$. Both antiferromagnets possess the perovskite crystal structure and remain basically cubic to the lowest temperatures. Furthermore, their magnetic properties have been investigated by a variety of techniques and most of the experimentally relevant coupling constants (magnetic, magnetoelastic, magneto-optic) have been determined experimentally. Despite this considerable knowledge, the low-temperature regime of NSLR appears to be somewhat puzzling and is not well understood.

$RbMnF_3$ has been previously studied by NMR (Refs. 3–5) and two relaxation regimes were identified. The high-temperature region ($0.05 \leq T/T_N \leq 1$; $T_N = 83$ K), strongly dependent upon temperature but independent of magnetic field, was also observed[6] in $KNiF_3$ and shown to be describable by a two-magnon Raman process effective in both systems.[7,8]

The ^{19}F low-temperature relaxation process in $RbMnF_3$ was studied in detail by Twerdochlib and Hunt,[5] who performed careful measurements of the magnetic-field dependence of T_1 in the temperature range $0.0012 \leq T/T_N \leq 0.04$ for magnetic field strengths $0.1 \leq H_0 \leq 0.5$ T. Their data indicated an approximately quadratic field dependence for most of the temperature range considered. This rather strong magnetic field dependence at such low field strengths was pointed out as being inconsistent with most mechanisms involving magnons. On the basis of these data, in Ref. 5 was proposed a new relaxation mechanism not involving the transferred hyperfine interaction between the ^{19}F nuclear spin and the electronic spin of the Mn^{2+} ion. Instead it was suggested that the Mn^{55} nuclei, quantized perpendicularly to the quantization axis of the ^{19}F spins, act as a bath and the ^{19}F spins relax to this bath via the ^{19}F-^{55}Mn nuclear magnetic dipole-dipole interaction.

The measurements presented here of the ^{19}F NSLR rates in isostructural $KNiF_3$ would enable us to test the validity of the relaxation model proposed in Ref. 5. They permit concluding that the partly unexpected magnetic field dependence of T_1 at low temperatures in these cubic quasi-isotropic antiferromagnets, is a consequence of a relaxation mechanism mediated by the transferred hyperfine interaction. Thus the behavior of T_1 as a function of Larmor frequency would probe the very low-frequency region of the spin-fluctuations in these magnetic systems. Since this region of frequency and of low temperatures is not readily accessible by other techniques, the interpretation of NSLR results in the present case may be of considerable interest in the understanding of the spin dynamics of a quasi-isotropic antiferromagnet for $T \ll T_N$.

In Sec. III we discuss the possibility that the puzzling relaxation mechanism prevalent at low temperatures may involve a diffusive behavior for the fluctuations in the magnetic energy.[9] Although the experimental data suggest that such process may be responsible for the Larmor frequency dependence of T_1 in $KNiF_3$ and $RbMnF_3$, a full quantitative description from a microscopic point of view is not available at the present time. The experimental results obtained in $KNiF_3$ are presented in Sec. II.

II. T_1 MEASUREMENTS IN $KNiF_3$

Using conventional pulsed NMR, we have measured the ^{19}F NSLR time T_1 in $KNiF_3$ for magnetic-field strengths $0.4 \leq H_0 \leq 3.0$ T and temperatures in the range $4.2 \leq T \leq 30$ K ($T_N = 246$ K). A crystal was aligned with a $\langle 100 \rangle$ direction parallel to the external field $\mathbf{H}_0$ and T_1 was measured separately for the two ^{19}F resonance lines.

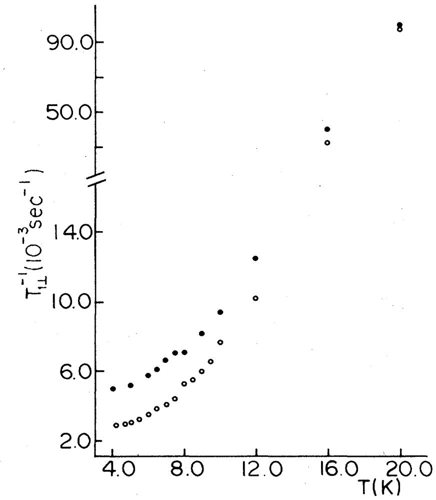

FIG. 1. $^{19}F_{\perp}$ spin lattice relaxation rate in $KNiF_3$ as a function of temperature for $H_0 || \langle 100 \rangle$. Fluorine Larmor frequencies are (●) $\nu = 19$ MHz and (○) $\nu = 54$ MHz. Notice the change of vertical scale above the break.

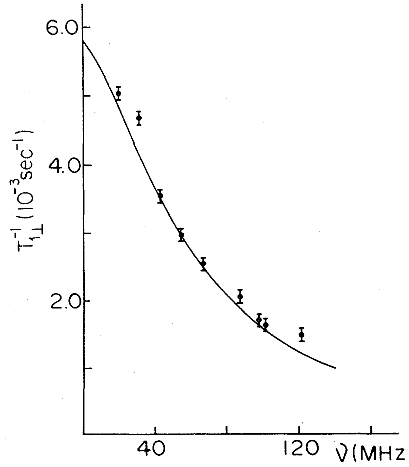

FIG. 2. $^{19}F_{\perp}$ spin lattice relaxation rate in $KNiF_3$ as a function of Larmor frequency for $H_0 || \langle 100 \rangle$ and $T = 4.2$ K. The solid line is a fit obtained from Eq. (7).

The high-field line corresponds to ^{19}F spins ($F_{\perp}$) whose F-Ni internuclear vector is perpendicular to H_0, while the low-field resonance ($F_{||}$) corresponds to a parallel orientation of the internuclear vector. For magnetic-field strengths $H_0 \geqq 0.7$ T we expect domain-wall movements to have ceased in our crystal and to encounter only two types of domains denoted by d_x and d_y.[10] These are domains where the sublattice magnetization is perpendicular to the applied field and oriented along the x and y axes, respectively ($H_0 || z$). In most of the range of magnetic-field strengths of our measurements, these two lines are sufficiently separated to prevent cross relaxation from taking place at rates comparable to $1/T_1$. It is then possible to measure independently the spin-lattice relaxation time T_1 and $F_{\perp}$ and $F_{||}$ nuclear spins in "flopped" domains. At high magnetic field strengths $H_0 \gtrsim 2.5$ T and low temperatures, T_1 becomes quite long in $KNiF_3$ (~ 12 min for $F_{\perp}$ at 4.2 K) becoming somewhat difficult to measure with accuracy.

Figure 1 shows the result of our $^{19}F_{\perp}$ NSLR rate measurements in $KNiF_3$ as a function of temperature for two values of the applied magnetic field parallel to a $\langle 100 \rangle$ crystal direction. For $T \lesssim 15$ K a magnetic-field dependent mechanism becomes important and finally dominates over the relaxation mechanism prevalent at higher temperatures. This high-temperature regime strongly temperature dependent and independent of magnetic field has been shown to be accountable by a two-magnon Raman process mediated by the transferred hyperfine interaction.[7] A very similar behavior with two clearly different relaxation regimes is also observed in $RbMnF_3$.[3–5] For $T \lesssim 10$ K to $KNiF_3$, the NSLR rate of $^{19}F_{\perp}$ spins denoted by $(1/T_1)_{\perp}$, can be seen from Fig. 1, to vary rather slowly with temperature. We assume that the contribution from the high-temperature relaxation process is negligible in this region.

Of special interest are the experimentally determined ratios $(1/T_1)_{\perp}/(1/T_1)_{||}$ as a function of temperature. We observed very little change of this ratio from $T = 4.2$ K to $T = T_N = 246$ K, despite the six orders of magnitude variation of T_1. In particular, there is no significant change in the ratio when one goes from the high-temperature relaxation regime to the field-dependent low temperature one.

The experimentally determined values of $(1/T_1)_{\perp}/(1/T_1)_{||}$ vary from 2.4 ± 0.2 to 3.4 ± 0.3 over the whole temperature range. This strongly suggests for both processes, a mechanism mediated by the F-Ni hyperfine interaction as discussed in Sec. III.

Figure 2 shows the result of our measurements of the Larmor frequency dependence of $(1/T_1)_{\perp}$ in $KNiF_3$ at $T = 4.2$ K. For frequencies $\nu \lesssim 20$ MHz we expect cross-relaxation between $^{19}F_{\perp}$ and $^{19}F_{||}$ spins to be significant so that the datum at $\nu = 20$ MHz in Fig. 1 should only be considered as a lower limit for $(1/T_{\perp})$. For comparison Fig. 3 shows the ^{19}F relaxation rate $1/T_1$ at $T = 0.1$ K in

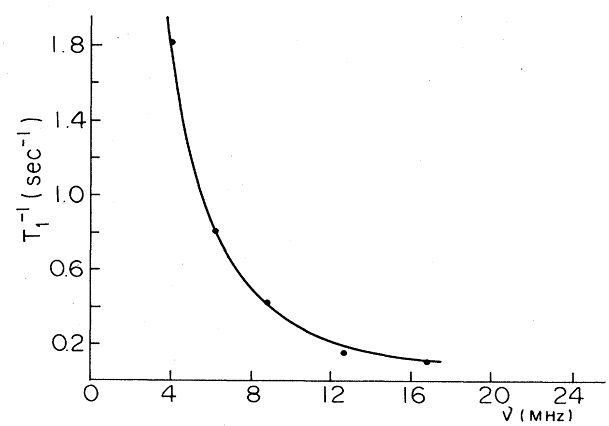

FIG. 3. Larmor frequency dependence of the ^{19}F spin lattice relaxation in $RbMnF_3$ at $T = 1.0$ K from the data of Twerdochlib and Hunt. The solid line is a fit obtained from Eq. (7).

$RbMnF_3$ as a function of frequency from the work of Twerdochlib and Hunt.[5] Instead of the quadratic frequency dependence prevailing in $RbMnF_3$ over a considerable temperature range, a slower, temperature-dependent decay of $(1/T_1)_\perp$ is observed in $KNiF_3$.

III. DISCUSSION

Our experimental results in $KNiF_3$ for $T \lesssim 0.04 T_N$ cannot be accounted for by the mechanism proposed in Ref. 5 for the low-temperature relaxation of isostructural $RbMnF_3$. This process, based upon the magnetic dipole-dipole interaction between the nuclear spins of the magnetic ions and the ^{19}F spins, should be very ineffective in $KNiF_3$ where 99% of nickel nuclei do not possess nuclear spin. Moreover, such a mechanism could not account for the relatively large ratio $(1/T_1)_\perp/(1/T_1)_\parallel \sim 3$ prevalent in $KNiF_3$ at all temperatures. The similarity in the behavior of $RbMnF_3$ and $KNiF_3$ further suggests that a common process may be responsible for the low-temperature regime of spin-lattice relaxation in either case.

It was pointed out in Ref. 5 that standard relaxation mechanisms involving magnons or magnons and phonons are not able to explain the observed magnetic field dependence of T_1 in $RbMnF_3$. The same observation is applicable to the present data in $KNiF_3$ for magnetic-field strengths small compared to $(2H_AH_E)^{1/2} \sim 4.5$ T.

The possibility that the ^{19}F spin-lattice relaxation mechanism in $KNiF_3$ and $RbMnF_3$ may involve a diffusive mode below T_N will be discussed in some detail. As predicted by the hydrodynamic theory[9] a diffusive mode is also expected in an isotropic antiferromagnet below T_N, in addition to spin waves. This mode should manifest itself in the correlation function of conserved quantities such as the magnetic energy and the component of magnetization parallel to the direction of antiferromagnetic ordering. The relaxation rate of a ^{19}F nucleus in $KNiF_3$ can be expressed in terms of correlation functions of such conserved quantities.

One can consider for example a $^{19}F_\perp$ spin in an antiferromagnetic domain where the direction ordering is along the y axis (d_y domain) with the external magnetic field applied along the z axis. Since the correlation function for components of spin transverse to the direction of antiferromagnetic ordering is dominated by magnons, we keep in the relaxation Hamiltonian[7] $\mathscr{H}_r$ only those terms which contain longitudinal components of spin:

$$\mathscr{H}_r = A_\parallel I_y (S_y^A + S_y^B) . \tag{1}$$

Here $\mathbf{S}_A$ and $\mathbf{S}_B$ refer to magnetic-ion spin operators on both sublattices. Each $^{19}F_\perp$ nuclear spin $\mathbf{I}$ in a d_y domain is assumed to be coupled through the component $A_\parallel$ of the hyperfine tensor, to two magnetic ions at sites separated by a crystal unit-cell distance a. The expression for $1/T_1$ can be obtained from the theory of Moriya[11] and expressed in the form

$$1/T_1 = (A_\parallel/\hbar)^2 \int_{-\infty}^{\infty} \cos(\omega t) [\langle\{\delta S_y^A(t);\delta S_y^A\}\rangle + \langle\{\delta S_y^A(t);\delta S_y^B\}\rangle] dt . \tag{2}$$

The curly brackets in Eq. (2) denote anticommutators while the angular brackets are thermal averages and $\delta\mathbf{S} = \mathbf{S} - \langle\mathbf{S}\rangle$. The time dependence of $\delta\mathbf{S}(t)$ is assumed to be governed by a Heisenberg exchange Hamiltonian and $\omega/2\pi = \nu$ denotes the Larmor frequency of the fluorine nuclear spins.

In Eq. (1) it is assumed that for a $^{19}F_\perp$ spin in a d_y domain, the F-Ni internuclear vector is parallel to the sublattice magnetization. If instead of a d_y domain we would have considered a $^{19}F_\perp$ spin in a d_x domain, the result for $(1/T_1)_\perp$ would differ from Eq. (2) only in the substitution of $A_\parallel$ by $A_\perp$. However for a $^{19}F_\parallel$ spin, the relaxation rate $(1/T_1)_\parallel$ would be given by Eq. (2) with the substitution of $A_\parallel$ by $A_\perp$, regardless of belonging to a d_x or d_y domain. This occurs because for a $^{19}F_\parallel$ spin, the F-Ni internuclear vector is always perpendicular to the sublattice magnetization in both types of flopped domains.

From Eq. (2) one can then derive an expression for the ratio $(1/T_1)_\perp/(1/T_1)_\parallel$. The relaxation rate of $^{19}F_\perp$ spins should depend upon the populations of ^{19}F spins in d_x and d_y domains. If they are approximately equal and a spin temperature is established, all $^{19}F_\perp$ spins are expected to relax at a common rate given by $(1/T_1)_\perp \propto \frac{1}{2}(A_\parallel^2 + A_\perp^2)$. On the other hand, the relaxation rate of $^{19}F_\parallel$ spins should be independent of the relative populations in d_x and d_y domains. If the $^{19}F_\parallel$ resonance line does not overlap appreciably with that of $^{19}F_\perp$ spins, one expects $(1/T_1)_\parallel \propto A_\perp^2$. Using the numerical values[12] $A_\parallel = 50.5\times10^{-4}$ cm^{-1} and $A_\perp = 25.6\times10^{-4}$ cm^{-1} for $KNiF_3$, we obtain a ratio $(1/T_1)_\perp/(1/T_1)_\parallel = \frac{1}{2}[1+(A_\parallel/A_\perp)^2] = 2.45$ in reasonable agreement with the experimental results of Sec. II. For $RbMnF_3$ this ratio is expected to be close to unity.

From Eq. (2) one can derive a more useful expression introducing spatial Fourier transforms

$$\delta S_{yq}^A = \frac{1}{\sqrt{N}} \sum_j \delta S_{yj}^A e^{i\mathbf{q}\cdot\mathbf{r}_{jA}} ,$$
$$\delta S_{yq}^B = \frac{1}{\sqrt{N}} \sum_j \delta S_{yj}^B e^{i\mathbf{q}\cdot\mathbf{r}_{jB}} , \tag{3}$$

and operators for the longitudinal components of magnetization δM_{yq}, and staggered magnetization δN_{yq}

$$\delta M_{yq} = \sum_{\alpha=A,B} \delta S_{yq}^\alpha ,$$
$$\delta N_{yq} = \sum_{\alpha=A,B} \epsilon_\alpha \delta S_{yq}^\alpha \quad \text{with } \epsilon_A = +1, \ \epsilon_B = -1 . \tag{4}$$

The summations in Eq. (3) are over all magnetic-ion sites of a given sublattice and N denotes the total number of magnetic unit cells.

Using Eqs. (3) and (4) together with a spherical approximation over a Brillouin zone of radius $q_{\max}=(3/4\pi)^{1/3}2\pi/a$, Eq. (2) can be written in the following form:

$$1/T_1=\tfrac{3}{2}(A_{\parallel}/\hbar)^2\frac{1}{q_{\max}^3}\left[\int_0^{q_{\max}} C_{m_y m_y}(q\omega)[q^2+(q/a)\sin(qa)]dq+\int_0^{q_{\max}} C_{n_y n_y}(q\omega)[q^2-(q/a)\sin(qa)]dq\right], \tag{5a}$$

where

$$C_{n_y n_y}(q\omega)=\tfrac{1}{2}\int_{-\infty}^{\infty}\cos(\omega t)\langle\{\delta N_{yq}(t);\delta N_{y-q}\}\rangle dt\ ,$$

$$C_{m_y m_y}(q\omega)=\tfrac{1}{2}\int_{-\infty}^{\infty}\cos(\omega t)\langle\{\delta M_{yq}(t);\delta M_{y-q}\}\rangle dt\ . \tag{5b}$$

The form of the longitudinal correlation functions $C_{m_y m_y}(q\omega)$ and $C_{n_y n_y}(q\omega)$ in an isotropic antiferromagnet has been investigated in great detail by several authors.[9,13–15] In the limit $q \ll q_{\max}$ the hydrodynamic theory[9] predicts that $C_{m_y m_y}(q\omega)$ should have a diffuse behavior below T_N. This prediction has been verified experimentally by neutron scattering experiments[16] in $RbMnF_3$. On the other hand, the staggered magnetization is not a conserved quantity in an isotropic antiferromagnet but is coupled to the magnetic energy, which is conserved. As a consequence it has been suggested by Halperin and Hohenberg[9] that the correlation function $C_{n_y n_y}(q\omega)$ should be the sum of two components, one a diffusive component, proportional to the correlation function for fluctuations in the magnetic energy and a second component representing the fast fluctuations of the nonconserved magnitude of the order parameter. This second component is expected to have a very broad spectrum and should not cause any frequency dependence in $1/T_1$. It will be assumed to be negligible and ignored in the following discussion. If one neglects the frequency-independent component, one can write the following expression[17,9] for the correlation function $C_{n_y n_y}(q\omega)$ for $q \ll q_{\max}$:

$$nC_{n_y n_y}(q\omega)\approx(1/\zeta^2)\left[\frac{\partial N_y}{\partial T}\right]^2 C_{\epsilon\epsilon}(q\omega)=\frac{2k_B T^2}{\zeta}\left[\frac{\partial N_y}{\partial T}\right]^2\frac{D_T q^2}{\omega^2+(D_T q^2)^2}\ , \tag{6}$$

where ζ and N_y are, respectively, magnetic-heat capacity and equilibrium staggered magnetization per unit volume, n is the number of magnetic ions per unit volume, and D_T is a diffusion coefficient for thermal conduction within the magnetic system. The coefficient $(1/\zeta)(\partial N_y/\partial T)^2$ can be calculated from spin-wave theory and compared with the corresponding coefficient of $nC_{m_y m_y}(q\omega)$ proportional to the parallel magnetic susceptibility $\chi_{\parallel}$.[9] It is found that at low temperatures $T \ll T_N$, the integral in Eq. (5a) containing the correlation function $C_{n_y n_y}(q\omega)$ makes the dominant contribution. From these arguments one can conclude that the relevant correlation function in Eqs. (5) appears to be that corresponding to magnetic energy fluctuations $C_{\epsilon\epsilon}(q\omega)$ given in Eq. (6). This enables one to write the following expression for the relaxation rate of a $^{19}F_{\perp}$ spin in a d_y domain

$$1/T_1\approx(3/2q_{\max}^3)(A_{\parallel}/\hbar)^2\frac{2k_B T^2}{\zeta}\left[\frac{\partial N_y}{\partial T}\right]^2\int_0^{q_c(T)}\frac{D_T q^2}{\omega^2+(D_T q^2)^2}[q^2-(q/a)\sin(qa)]dq\ . \tag{7}$$

We have introduced in the upper limit of the integral in Eq. (7) a cutoff wave vector $q_c(T) \ll q_{\max}$. Although this admittedly crude expedient is being adopted mainly for heuristic reasons it is not inconsistent with the ideas proposed by Michel and Schwabl.[15,18] These authors suggested that the diffusive behavior in $C_{\epsilon\epsilon}$ is only expected for $C_\theta q < \omega_i$. C_θ is a temperature-dependent second magnon velocity and ω_i is a relaxation frequency for scattering of thermally excited magnons, either by impurities at low temperatures or by umklapp processes at higher temperatures. For larger wave vectors the height of the central peak is expected to shift to the wings giving rise, under special window conditions, to a propagating mode (second magnon). For ferromagnetic linear chains some of these ideas have been recently confirmed by molecular dynamics calculations.[19]

Although it is not possible from our NMR data to test the validity of the second magnon concept in the system studied, we will nevertheless adopt a cutoff wave vector $q_c(T)$ in Eq. (7) and treat it as an adjustable parameter. For moderately pure, defect-free crystals one would then expect[18] a very small cutoff $q_c \ll q_{\max}$.

The mechanism we are discussing can only explain the experimental results of Sec. II if in the temperature range of interest $D_T q^2 \lesssim \omega$ for $q \leq q_c(T)$. This implies that the diffusive central peak in the correlation function for magnetic energy fluctuations must become extremely narrow in $KNiF_3$ and $RbMnF_3$, otherwise no significant Larmor frequency dependence could be obtained from Eq. (7). Although no reliable estimate of the diffusion coefficient D_T or its temperature dependence appears to be available presently, the possibility of such a narrow peak in $C_{\epsilon\epsilon}(q\omega)$ is strongly suggested by recent experimental results in $KNiF_3$. Light scattering in the temperature range 150 K $\lesssim T \lesssim T_N$ and wave numbers $q \sim 0.22\times 10^{-2}(\pi/a)$, attributed to magnetic energy fluctuations in $KNiF_3$,[20] has revealed a very narrow central peak whose width was estimated to be less than 600 MHz. At much lower temperatures it appears to be plausible that this width may reach, for $q \lesssim q_c(T)$, the range $\omega/2\pi \sim 100$ MHz characteristic of the ^{19}F Larmor frequencies of our experiments. This could explain the observed frequency dependence of $1/T_1$ predicted by Eq. (7) and also support the view that the origin of the proposed ^{19}F spin-lattice relaxation mechanism may be found in the hydrodynamic fluctuations of the magnetic energy.

From Eqs. (6) and (7) one can compute the nuclear spin-lattice relaxation rates and compare with the experimental results. The coefficients of Eq. (6) can be calculated from spin-wave theory.[21,22] In the temperature range $(2H_AH_E)^{1/2}\gamma h/k_B \lesssim T \ll T_N$ one finds

$$2k_BT^2(\partial N_y/\partial T)^2/\zeta = (96.2/\pi^4)(T/\Theta_N) ,$$

where $\Theta_N = \gamma h H_E q_{max} a/\sqrt{2}k_B$.

Figure 2 shows a fit of the experimental spin lattice relaxation rates for $^{19}F_\perp$ spins in $KNiF_3$ as a function of Larmor frequency resulting from Eq. (7). Good agreement with the non-Lorentzian frequency dependence observed at 4.2 K is obtained for $q_c/q_{max} = 1.69\times 10^{-2}$ and $D_Tq^2_{max} = 2.19\times 10^{12}$ sec^{-1}. For an isotropic antiferromagnet this value of the cutoff wave vector is consistent with the assumption of a hydrodynamic regime implicit in Eq. (7). As expected, q_c is much smaller than q_{max} but it is still larger than a minimum wave vector[9] $q_0 = (4/3\pi^2)^{1/3}(3H_A/2H_E)^{1/2}$. For $q \lesssim q_0$ the magnon dispersion relationship deviates from a linear q dependence due to the effects of anisotropy. For $KNiF_3$, $q_0/q_{max} \sim 0.5\times 10^{-2}$; while for $RbMnF_3$, $q_0/q_{max} \sim 0.13\times 10^{-2}$. Moreover, the width of the peak in $C_{\epsilon\epsilon}$ at the cutoff wave vector $D_Tq_c^2 = 6.2\times 10^8$ sec^{-1} is also believed to be compatible with the very narrow central peak observed by light scattering in $KNiF_3$.[20]

For $RbMnF_3$ the data shown in Fig. 3 exhibit a $1/\nu^2$ dependence in $1/T_1$. This is of course predicted by Eq. (7), provided $\nu > D_Tq_c^2/2\pi$ as shown by the fit of Fig. 3 for $T = 1.0$ K. However, from this fit to the $RbMnF_3$ data one can only determine upper and lower limits for the width and cutoff wave vector, respectively. The results are $D_Tq^2_{max} \le 1.3\times 10^{10}$ sec^{-1} and $q_c/q_{max} \geqq 0.047$ with $D_Tq_c^2(q_c/q_{max})^5 = 6.43$ sec^{-1}.

The large difference in the value of $D_Tq^2_{max}$ for $KNiF_3$ and $RbMnF_3$ at approximately the same value of T/Θ_N is somewhat intriguing. D_T is expected to be proportional to a typical relaxation frequency for quasimomentum-conserving scattering among thermally excited magnons.[18] Although this would imply a larger value of D_T for $KNiF_3$ because of the larger exchange field $[H_E(KNiF_3)/H_E(RbMnF_3) \approx 4]$, the very large difference in the values of D_T obtained from the fits could probably not be attributed solely to exchange interactions. On the other hand, the anisotropy field is much smaller in $RbMnF_3$ than in $KNiF_3$ $[H_A(KNiF_3)/H_A(R_bMnF_3) \sim 56]$ and could make a significant difference. However, the effect of anisotropy upon the thermal-diffusion coefficient of a magnetic system at very low temperatures is not well understood and needs to be elucidated.

IV. CONCLUSIONS

We conclude that the rather puzzling magnetic-field dependence of the ^{19}F spin lattice relaxation in $KNiF_3$ at low temperatures originates in a mechanism mediated by the transferred hyperfine interaction. Some similarities with the behavior of isostructural $RbMnF_3$ suggest that this may also be true in this system, unlike previous suggestions. The theoretical prediction of a central peak in the correlation function for hydrodynamic fluctuations in the magnetic energy is capable of describing the observed Larmor frequency dependence of $1/T_1$. The width of this peak obtained from spin lattice relaxation data is compatible with recent light scattering results in $KNiF_3$. The frequency dependence of the relaxation rate would then enable one to probe the very small q and small ω region of the spin fluctuations in a quasi-isotropic antiferromagnetic at low temperatures. This region of frequency, wave vector, and temperature is not readily accessible by other techniques.

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