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OBSERVATION OF TWO INTRINSIC NUCLEAR RELAXATION RATES IN ANTIFERROMAGNETIC $KMnF_3$

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This paper reports the results of the measurement of nuclear spin-lattice relaxation rates in $KMnF_3$, which indicate that there are at least two distinct relaxation processes in the ordered state. The high-temperature rate has a power-law temperature dependence, while the low-temperature rate has an approximate $\exp(-\alpha/T)$ dependence, indicating the effect of a magnon energy gap.

The first measurement[1] of relaxation times in $CuCl_2 \cdot 2H_2O$ led to the theoretical work of Moriya,[2] Van Kranendonk and Bloom,[3] and Mitchell.[4] These papers treated a magnon-Raman relaxation process and yielded the result (neglecting spin-wave interactions, using the long-wavelength limit, and assuming quantization of the electronic and nuclear spins along different directions in the crystal) that the nuclear relaxation rates should be proportional to T^3 for temperature above T_{AE}, and proportional to $T^2 \exp(-T_{AE}/T)$ for temperatures below T_{AE}, where kT_{AE} is the width of the magnon energy gap. Most experimental data have been in poor agreement with these predictions.

Pincus and Winter[5] have described a mechanism by which thermal phonons can participate directly in the nuclear relaxation process. This mechanism, applicable only when $T \ll T_{AE}$, yields a linear temperature dependence for the direct process and a T^7 dependence for the Raman process. In $CuCl_2 \cdot 2H_2O$, where $T_{AE} \approx 1°K$, an approximate T^7 dependence has been observed[6,7] between 1.25 and 0.95°K.[7] However, in this temperature region the Pincus-Winter theory is not valid, and, even if it were, the magnitude of the coupling is too small to explain the experimental results. In this temperature range for $T > T_{AE}$, a pure spin-wave process should predominate in this crystal. Abkowitz and Lowe[8] also observed a T^7 temperature dependence for protons in $CoCl_2 \cdot 6H_2O$ between 2 and 1.24°K but again these measurements are in the temperature range where a pure spin-wave process is expected.

Recently, Pincus[9] and Narath and Fromhold[10] have described a three-magnon process which has been observed in $CrCl_3$.[10] In this material, the Cr^{53} hyperfine interaction is almost isotropic, resulting in the exclusion of a magnon-Raman process to first order. The three-magnon process in the ferromagnetic state was investigated both theoretically and experimentally as a function of magnetic field for two external magnetic field directions, and there is reasonable agreement between theory and experiment. The temperature dependence for the three-magnon process has been calculated by Pincus[9] and found to be T^5 for $T > T_{AE}$, and to be proportional to a product of terms, one of which is $\exp(-T_{AE}/T)$, below T_{AE}. This exponential dependence is characteristic of any process influenced by a gap in the magnon spectrum.

Finally, the only good experimental evidence of the effect of an energy gap in an antiferromagnet was given by Kaplan et al.,[11] in MnF_2, where they measured the angular and temperature dependences of the relaxation rate of F^{19}. They reported excellent agreement with a theoretical calculation of the coupling of the nuclear spin to the magnons through the transferred hyperfine interaction. The result of the calculation shows that the low temperature relaxation rate falls off faster with decreasing temperature than the $T^2\exp(-T_{AE}/T)$ dependence predicted in the original work.[2]

The relaxation times of the F^{19} nuclei at the two nonequivalent sites in two different samples of $KMnF_3$ (denoted by samples 5 and 7) were measured using the spin-echo technique to observe the exponential recovery of the F^{19} nuclear magnetization following saturation by a series of rf pulses. The temperature range investigated was 1.25 to 25°K. Standard helium vapor-pressure techniques were used to measure the temperature below 4.06°K, and a calibrated germanium thermometer was used above. The relaxation rates are given in Figs. 1 and 2. The following points should be noted from these two figures. (1) Above 12°K, both samples 5 and 7 follow a power-law temperature dependence: sample 5, a T^5 and sample 7, a T^7. (2) Both samples break from this power-law dependence at approximately 12.5°K, both show a somewhat similar $\exp(-\alpha/T)$ below 12.5°K, and both have approximately the same relaxation rates between 3.5 and 12.5°K.

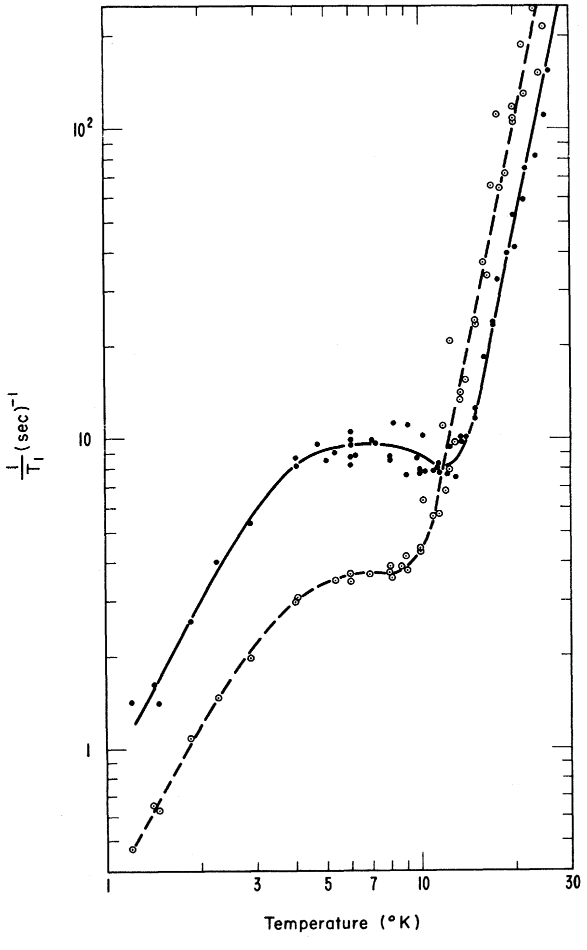

FIG. 1. F^{19} spin-lattice relaxation rates for nuclei at two inequivalent sites in $KMnF_3$ (sample 5). The circles are data from the high-field site (approximately 16 MHz) and the dots from the low-field site (approximately 14.7 MHz).

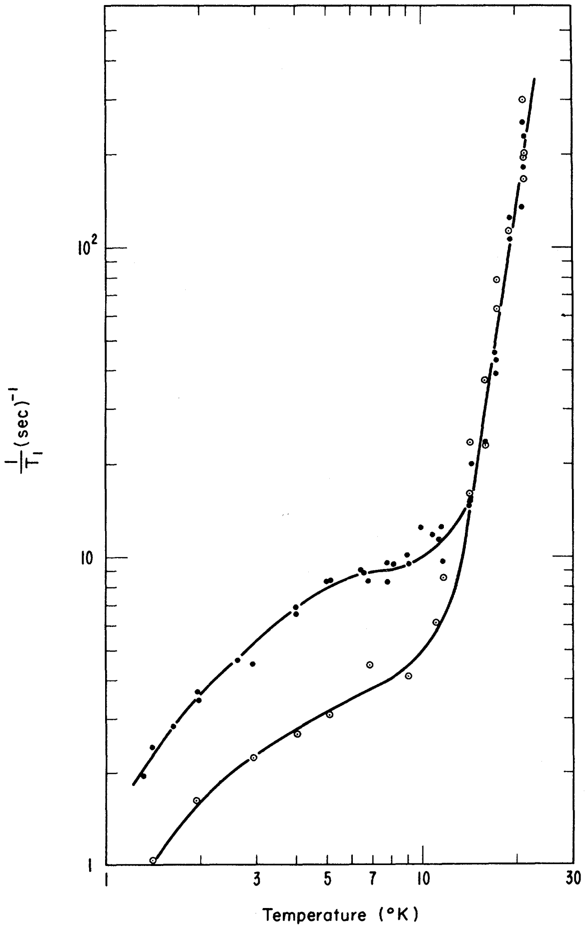

FIG. 2. F^{19} spin-lattice relaxation rates for nuclei at the two inequivalent sites in $KMnF_3$ (sample 7). The circles are data from the high-field site and the dots from the low-field site.

(3) The temperature dependence below 3.5°K is stronger in sample 5 than in sample 7.

$KMnF_3$ is a canted antiferromagnet over the temperature range investigated and has been described recently by a four-sublattice model by Minkiewicz and Nakamura.[12] Using this model, it is possible to calculate a T_{AE} of 0.3°K. It may be fortuitous, but $KMnF_3$ is the only antiferromagnet investigated thus far where the nuclear relaxation rate exhibits more than one intrinsic rate. Theoretically, the intrinsic relaxation rate for a magnetic material with a magnon energy gap should contain an exponential factor $\exp(-T_{AE}/T)$ for the temperature range $T<T_{AE}$. Experimentally, this rate has been observed[11,13] in two materials where this low-temperature dependence has predominated to temperatures above T_{AE}. The theoretical result implies that a power-law temperature dependence below T_{AE} is an impurity-dominated or nonintrinsic process. These nonintrinsic processes are independent of T_{AE} and will dominate the relaxation rate throughout the temperature range where the nuclear spin-impurity coupling is stronger than the nuclear spin-magnon coupling. There has been no theoretical or detailed experimental work on impurity relaxation processes in magnetic materials, so it is difficult to speculate on the form of their temperature dependences. However, a T^7 dependence has been observed[6-8] in a temperature region where one might expect an intrinsic exponential-dominated magnon rate. Prefaced with these remarks, it seems plausible that the low-temperature approximately exponential dependence, and the high-temperature T^5 dependence observed in $KMnF_3$ sample 5, are intrinsic rates, and the T^7 dependence is an impurity-dominated rate. This conclusion is further substantiated by the fact that sample 5 is known to have an impurity content less than 30 ppm (parts per million) and sample 7 could have an impurity content as large as 10 000 ppm.

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CLUSTER STRUCTURE OF EXCITED LEVELS IN He^6 AND Li^6†

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The light nuclei He^6 and Li^6 have received a great deal of attention both theoretically[1-3] and experimentally.[4-6] In particular, the states with excitation energies (E_x) less than 2 MeV in He^6 and the states with excitation energies less than 6 MeV in Li^6 are known to have predominantly a cluster structure of an alpha cluster plus a two-nucleon cluster in triplet or singlet s state.[3] In this communication, we report the result of a calculation using the resonating-group method[7,8] which shows that there also exist levels in He^6 with $E_x>2$ MeV which have predominantly a H^3-plus-H^3 cluster structure and levels in Li^6 with E_x between 3.5 and 10.5 MeV which have predominantly a H^3-plus-He^3 cluster structure.

Experimentally, there is some, but not definite, evidence[5,6] that there exist levels at 3.4 and 6.0 MeV in He^6 and a group of levels in the range from 6 to 10 MeV in Li^6. If these