DOUBLE RESONANCE AND NUCLEAR COOLING IN AN ANTIFERROMAGNET*

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We wish to report the observation of the Mn^{55} nuclear resonance by a double-resonance technique in the antiferromagnet $KMnF_3$. In addition, by observing the nuclear spin temperature as a function of power absorbed by the electronic antiferromagnetic resonance, we have discovered an apparent cooling of the nuclei via their interactions with the electron system.

Examination of the antiferromagnetic resonance of $KMnF_3$ in the liquid helium temperature range shows that even at these low temperatures there is a strong temperature dependence of the resonance field as is shown for one of the absorption lines by the experimental points of Fig. 1. We have interpreted this shift to be the result of a strongly temperature-dependent anisotropy field arising from the hyperfine interaction of the antiferromagnetic system with the Mn^{55} nuclear moments. This hyperfine anisotropy field has the form

$$H_{A_1} = (A/g\mu_B)\langle I_z\rangle.$$

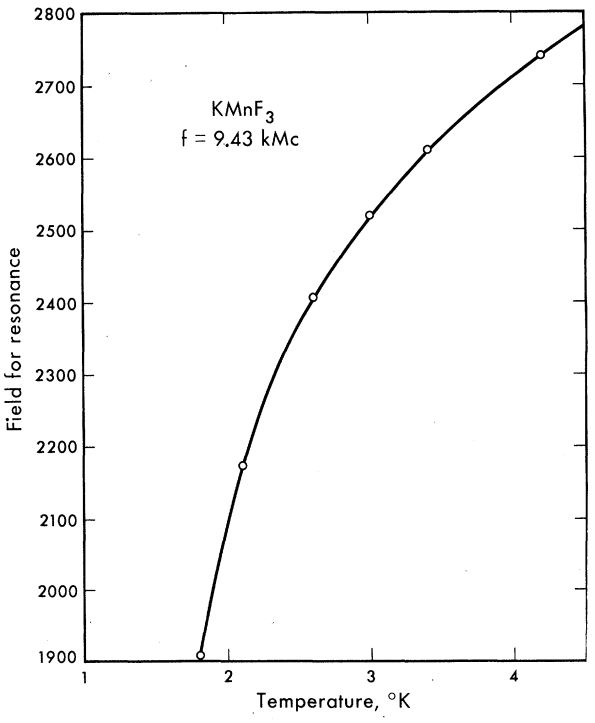

FIG. 1. Theoretical fit of the temperature dependence of the resonance field. The mechanism is the temperature dependence of the hyperfine anisotropy field.

The hyperfine constant $A/g\mu_B$ has been evaluated experimentally by Ogawa[1] from the paramagnetic resonance of Mn^{2+} in isomorphous compounds. Substituting Ogawa's value of 92 oe, we obtain

$$H_{A_1} = 8.0/T \text{ oe}.$$

The expression for the resonance field of the electronic antiferromagnetic resonance is

$$H_0 = -\tfrac{5}{2}H_{A_2} + [\tfrac{9}{4}H_{A_2}^2 + (\omega/\gamma)^2 + 5H_E H_{A_4} - 2H_E H_{A_1}]^{1/2}.$$

The anisotropy field H_{A_2} is responsible for the canting in this material[2] and is equal to 1300 oe as determined from measurements of the magnitude of the weak moment. The field H_{A_4}, which is associated with the cubic and axial anisotropy,[3] and the exchange field, H_E, were determined as 3.6 oe and 1.6×10^6 oe, respectively, by fitting theory and experiment. From the above expression it may be seen that the shift in resonance field resulting from the nuclear magnetization is of the order of $(H_E/H_0)H_{A_1}$. Thus a one-oersted nuclear field may produce a shift in resonance field by nearly one kilo-oersted. The completely satisfactory agreement between the experimental observations and this theory is shown in Fig. 1.

The strong dependence of the antiferromagnetic resonance field on nuclear spin temperature suggested the possibility of observing the Mn^{55} nuclear resonance in antiferromagnetic $KMnF_3$ by a double-resonance technique. If enough rf power is supplied at the nuclear resonance frequency to saturate the nuclear magnetization, a shift of the antiferromagnetic resonance to its high-temperature position is expected. From the measurements of Ogawa the expected nuclear resonance frequency is

$$\nu_0 = (A/h)\langle S\rangle = 630 \text{ Mc/sec}.$$

The sample was a small cube of $KMnF_3$, approximately 1 mm on a side, at the center of a small rf coil, slightly over 1 mm in diameter. The sample and coil were placed at the bottom of a rectangular microwave cavity resonating in a TE_{101} mode. A shift in the position of the antiferromagnetic resonance with rf power could be observed over a 42-Mc/sec range in frequency centered around 628 Mc/sec, in good agreement with the expected central frequency. In Fig. 2 is shown a

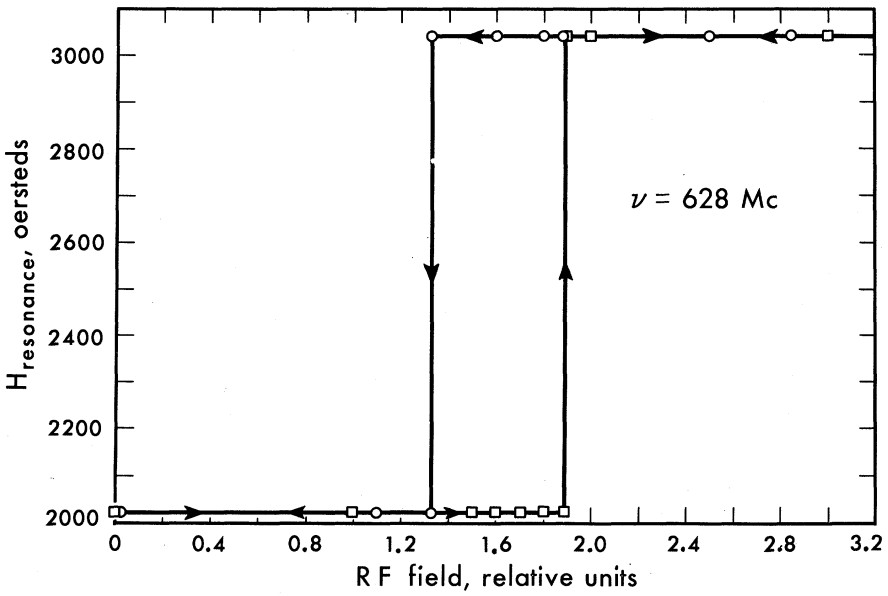

FIG. 2. Location of the antiferromagnetic resonance at 2.0°K as a function of rf field at 628 Mc/sec. The shift arises from the saturation of the nuclear magnetization.

plot of the location of the antiferromagnetic resonance as a function of rf field at the central frequency. It is found that when the rf field exceeds a critical value the electronic resonance line shifts abruptly to a new position characteristic of a high nuclear temperature. On reducing the rf field we find that the resonance line remains at its high-field position down to rf power levels well below those required for the initial shift.

By pulsing the 628-Mc/sec rf field on and then off, it was possible to observe the recovery of the position of the antiferromagnetic resonance line after removing the rf power. Such a recovery curve after nuclear saturation at 2.1°K is shown in Fig. 3. The relaxation appears to be nearly exponential with a characteristic relaxation time of 50 msec.

The very large shifts indicate that the nuclear magnetization is nearly reduced to zero at the rf levels employed. We estimate the interaction of the Mn^{55} nuclei via spin waves[4] to yield a characteristic time T_2 of the order of 0.15 μsec. With the observed T_1 of about 50 msec, the rf field for partial saturation should be approximately 2 oe. This is actually about the level at which the nuclear saturation jumps to nearly its full value. As is shown in Fig. 2 the nuclear saturation may actually be maintained at rf fields well below this value. We have analyzed the excitation of the Mn^{55} nuclei in some detail in an effort to understand the complex behavior observed. We find that because of the large parallel susceptibility of a canted antiferromagnet,[5] the Mn^{55} nuclear resonance is excited indirectly by the sublattice magnetization rather than directly by the rf field. However, the Mn^{55} nuclei themselves very strongly inhibit the sublattice excitation in the vicinity of the nuclear resonance as long as there is a substantial nuclear moment. This theory indicates that it should be harder to produce an initial saturation of the nuclei against their dynamical resistance than to maintain the saturation when there is very little

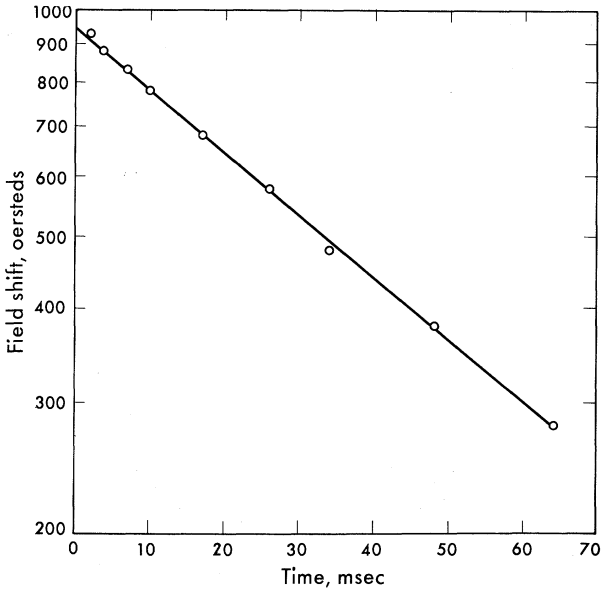

FIG. 3. Recovery curve showing the shift of the antiferromagnetic resonance at a given time after turning off the 628-Mc/sec rf power.

nuclear moment, in agreement with the observed hysteresis shown in Fig. 2. The frequency range over which such behavior may be expected depends on the nuclear magnetization and bears very little relation to the actual linewidth $1/T_2$.

Experiments have also been performed[6] in which the antiferromagnetic resonance is driven at high microwave levels without any direct nuclear excitation. Under these conditions we also find a shift in the position of the antiferromagnetic resonance. This shift is clearly associated with a change in nuclear magnetization. The apparent nuclear temperature as a function of microwave power for several lattice temperatures is shown in Fig. 4. At 4.2°K the nuclear temperature remains constant up to the power at which spin-wave breakdown occurs.[6] At high power levels the increased nuclear temperature presumably reflects the general increase in the level of excitation of the magnetic system. At 2.1 and 1.8°K there is a quite unexpected reduction in the nuclear temperature before the onset of spin-wave heating. At these lower temperatures and low power levels the nuclei are evidently cooled through their interactions with the off-equilibrium spin-wave system. The cooling of nuclear spins may be related to the possible development of an Overhauser effect in ordered magnetic materials as discussed by Oguchi and Keffer.[7] It should be emphasized that although the present experiments are unusually sensitive to a change in nuclear temperature, they are not especially sensitive to the development of a uniform nuclear polarization. Such a polarization would shift the resonance in the same way as an external field. Full nuclear polarization would produce a shift of only a few oersteds and would be completely dominated by the shift resulting from even a slight change in nuclear temperature. The recovery of the position of the antiferromagnetic resonance following microwave saturation has been studied. The relaxation appears to be the same as that observed following direct rf saturation of the nuclei.

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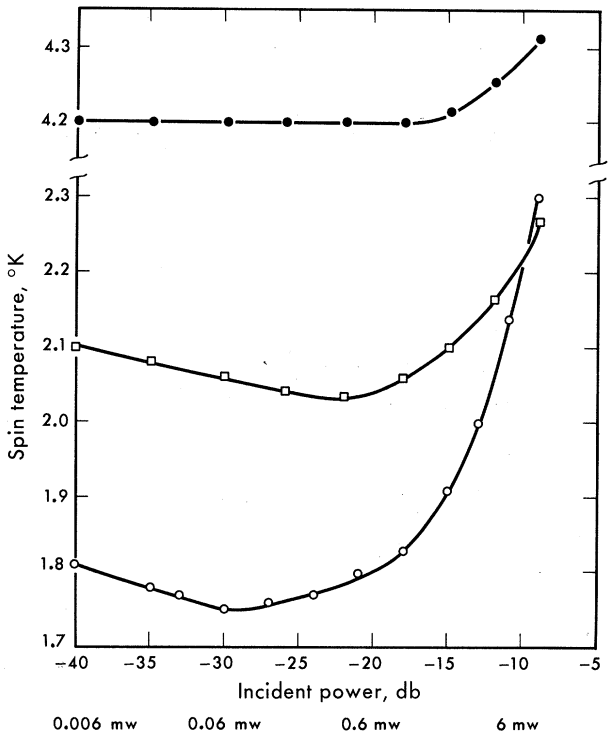

FIG. 4. Spin temperature as a function of microwave power absorbed by the sample. The spin temperature is determined by the resonance field.

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