# DISCOVERY OF A SIMPLE CUBIC ANTIFERROMAGNET: ANTIFERROMAGNETIC RESONANCE IN $RbMnF_3$

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Low-temperature magnetic-resonance measurements on $XMnF_3$ compounds have been briefly reported.[1] Of these perovskite-like structures ($X$ = Na, K, Rb, Cs) the most novel and interesting is $RbMnF_3$, because it is a simple cubic antiferromagnet. In contrast to the simple behavior of $RbMnF_3$, the other compounds of this series depart from cubic structure[2] and display weak ferromagnetism resulting from canting of the sublattice magnetizations.[3]

Cubic $RbMnF_3$ is characterized by a strong exchange interaction ($T_N \simeq 54.5°$) and small anisotropy; it is therefore an extremely attractive material for the study of antiferromagnetism. In this note we shall report theory and observation of antiferromagnetic resonance.

In Fig. 1 is shown the field for antiferromagnetic resonance as a function of angle for the applied field in a {100} plane and a microwave frequency of 23.285 kMc/sec. The fourfold symmetry indicates that the sublattice magnetizations are pulled away from the easy axis by the external field, since otherwise twofold symmetry would be observed even for cubic anisotropy. This is in agreement with static susceptibility measurements made by the force-balance method in which we found the susceptibility to be isotropic for fields above 5 kOe. The appearance of an antiferromagnetic resonance at fields greater than $\omega/\gamma$, the field for paramagnetic resonance, also indicates the presence of spin flopping. This can be seen from the resonance conditions given by Nagamiya, Yosida, and Kubo[4] for the case of a flopped uniaxial antiferromagnet with the applied field parallel to the anisotropy axis. In the uniaxial case the spin axis lies along an extremal of anisotropy energy, and the resonance condition is $H = [(\omega/\gamma)^2 + 2H_E H_A]^{1/2}$.

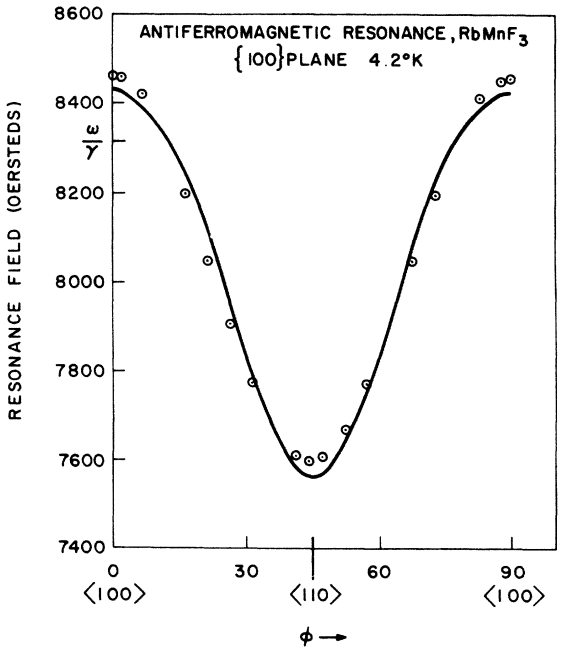


FIG. 1. Antiferromagnetic resonance at 23.285 Gc/sec for the applied field in a {100} plane.

The appearance of the minimum resonance field in the ⟨110⟩ direction, and the supplementary observation of an isotropic resonance when the applied field is in the {111} plane, indicate that the ⟨111⟩ direction is the easy axis. The resonance excited when the applied field is in the {111} plane is given by the usual resonance condition for the applied field perpendicular to the anisotropy axis, $H = [(\omega/\gamma)^2 - 2H_E H_A]^{1/2}$. We thus consider an anisotropy energy of the form $E_A = K(\alpha_1^4 + \alpha_2^4 + \alpha_3^4)$ for each sublattice, where $K$ is positive and the $\alpha$'s are the direction cosines of the magnetization with respect to the crystalline axes. When the spins are near an easy direction, the anisotropy may be related to an effective field in the ⟨111⟩ direction given by $H_A = 8K/3g\mu_B S$, where $K$ is the anisotropy energy per ion.

For an applied field greater than $H_C$ in an arbitrary direction, the spin axis lies in the plane perpendicular to the field, with the sublattice magnetization tipped into the direction of the field by a small angle, $\chi = H_0/2H_E$, and the position of the spin axis in the plane located at the minimum of anisotropy energy within the plane.

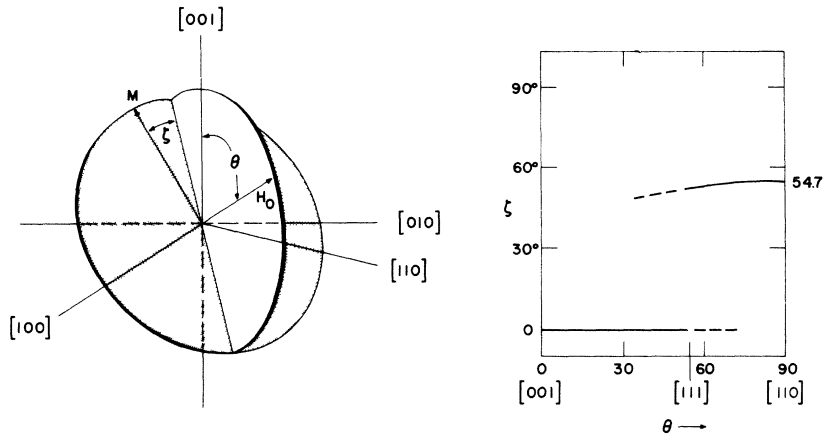


FIG. 2. Illustration of the spin configuration for the applied field in a {110} plane when the field is greater than the critical field, $(2H_EH_A)^{1/2} = 2450$ Oe.

The situation is illustrated in Fig. 2 for the case of the applied field in the {110} plane.

To complete the basis of the resonance theory we must include the hyperfine anisotropy field. This field, arising from the polarization of $Mn^{55}$ nuclear moments, gives rise to the strong temperature dependence of the antiferromagnetic-resonance field at low temperatures which was first observed in $KMnF_3$.[5] It is introduced in the theory as an effective field, $H_N$, in the direction of the sublattice magnetization as determined above. The value of the hyperfine anisotropy field, $H_N = (A/g\mu_B)\langle I_z\rangle$ may be obtained from precise electron paramagnetic resonance measurements[6] of $A^{55}$ in $KMgF_3$:$Mn^{2+}$ corrected for volume differences between the diamagnetic host and the concentrated $RbMnF_3$ lattice. The result is $H_N = 9.43/T$ Oe.

The equation of motion for each sublattice may be written

$$(1/\gamma)d\vec{M}/dt = \vec{M}\times(\vec{H}_E + \vec{H}_0 + \vec{H}_N) + \vec{T},$$

where $\vec{T}$ is the torque arising from the anisotropy, and is given by

$$T_x = \sin\phi_m\,\partial E_A/\partial\theta_m + \cot\theta_m\cos\phi_m\,\partial E_A/\partial\phi_m,$$

$$T_y = -\cos\phi_m\,\partial E_A/\partial\theta_m + \cot\theta_m\sin\phi_m\,\partial E_A/\partial\phi_m,$$

$$T_z = -\partial E_A/\partial\phi_m,$$

where $\theta_m$ and $\phi_m$ are the polar angles of the sublattice magnetizations. We transform to new coordinates $x', y', z'$ and $x'', y'', z''$, where $z'$ and $z''$ are along the equilibrium directions of $\vec{M}_1$ and $\vec{M}_2$, the sublattice magnetizations, and $x'$ and $x''$ are perpendicular to $\vec{H}_0$. The equations of motion become

$$\frac{1}{\gamma}\frac{d}{dt}\begin{pmatrix} M_1^{x'} \\ M_1^{y'} \\ M_2^{x''} \\ M_2^{y''} \end{pmatrix} = \begin{pmatrix} 0 & a & 0 & b \\ -b & 0 & c & 0 \\ 0 & b & 0 & a \\ c & 0 & -a & 0 \end{pmatrix}\begin{pmatrix} M_1^{x'} \\ M_1^{y'} \\ M_2^{x'} \\ M_2^{y'} \end{pmatrix} + \begin{pmatrix} \delta T_1^{x'} \\ \delta T_1^{y'} \\ \delta T_2^{x''} \\ \delta T_2^{y''} \end{pmatrix},$$

where $a = (H_E + H_N)$, $b = (-H_E + H_0^2/2H_E)$, $c = -H_E$, and $\delta\vec{T}_1$ and $\delta\vec{T}_2$ are the changes in torque for small displacements of $\vec{M}_1$ and $\vec{M}_2$ from their "flopped" equilibrium positions.

We now calculate the resonance frequency and obtain

$$(\omega/\gamma)^2 = H_0^2 + 3B(\theta,\phi)H_EH_A + 2H_EH_N,$$

where $\theta$ and $\phi$ are the polar and azimuthal angles

of $\vec{H}_0$. The angular dependence is given by

$$B(\theta, \phi) = 3(\alpha_1^2 l_1^2 + \alpha_2^2 l_2^2 + \alpha_3^2 l_3^2) - (\alpha_1^4 + \alpha_2^4 + \alpha_3^4),$$

where $\alpha_i$ is a direction cosine of the equilibrium spin axis and $l_i$ is a direction cosine of $\vec{H}_0$. For the applied field in the (001) plane, the angle between the sublattice magnetization and the [001] axis is given by $\theta_m = \cos^{-1}(3+\cos 4\phi)/(7+\cos 4\phi)$, and the angular function in the resonance equation is

$$B(\tfrac{1}{2}\pi, \phi) = -4\cos 4\phi/(7+\cos 4\phi).$$

For the applied field in the (011) plane, the orientation of the magnetization is shown in Fig. 2. The configuration of lowest energy is shown by the solid line and the dashed lines are the extensions of these solutions beyond their stable range. (When the field is perpendicular to the {111} plane, the spins are free to rotate in the plane.) The angular function in the resonance equation is

$$B(\theta, \tfrac{1}{4}\pi) = -1 + \tfrac{13}{2}\cos^2\theta - 6\cos^4\theta,$$

for $\theta$ between [001] and [111], and

$$B(\theta, \tfrac{1}{4}\pi) = (2 - \sin^2\theta)(3\sin^2\theta - 1)/(2 + \sin^2\theta)$$

for $\theta$ between [111] and [011]. In Fig. 3 we show the excellent agreement between theory and experiment. For the applied field along [111], a single resonance is observed. However, for a few degrees to either side, a second resonance is observed. It can be shown that the slight misalignment out of the {110} can permit the formation of metastable domains having spin configuration with the resonance frequencies calculated for the extensions shown in Fig. 2. The intensity of the second resonance falls off rapidly with angle away from [111] as the formation of these domains becomes more energetically unfavorable.

Using the calculated value of $H_N$ we obtain $H_E = (890 \pm 20)$ kOe and $H_A = (4.47 \pm 0.04)$ Oe by a fit to the experimental data. The experimental value of $H_E$ is influenced by systematic error, while the smaller error assigned to $H_A$ arises from the uncertainty in $\langle S \rangle$. Both the sign and the magnitude of the anisotropy suggest that the principal mechanism is cubic crystal field splitting. The relation between $K$ and $a$, the crystal field parameter, is[7] $K = S(S-\frac{1}{2})(S-1)(S-\frac{3}{2})(a/6) = 5/4a$, and we obtain $K = 3.92\times10^{-4}$ cm$^{-1}$, while the measurements of Ogawa[8] give values of $a \sim 4\times10^{-4}$ cm$^{-1}$. The zero-point dipolar anisotropy has the opposite sign and may be calculated from Pearson's[9] formula to be $\sim 0.25\times10^{-4}$ cm$^{-1}$.

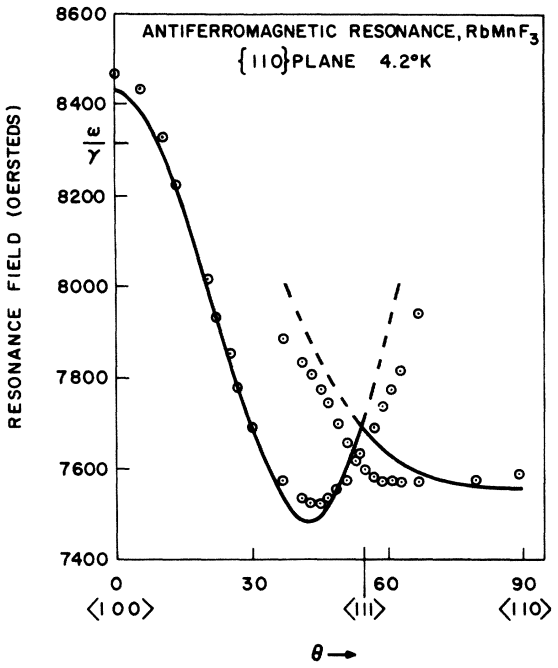


FIG. 3. Antiferromagnetic resonance at 23.285 Gc/sec for the applied field in a {110} plane.

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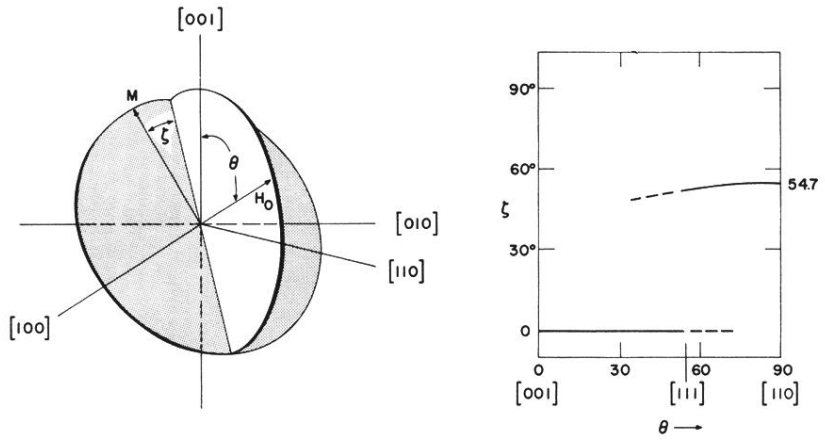


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