Antiferromagnetic Domains in $RbMnF_3$ †

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The indirect observation of domains in the cubic antiferromagnet $RbMnF_3$ through antiferromagnetic resonance is reported. Because of the cubic anisotropy of $RbMnF_3$, the equilibrium orientation of the magnetic sublattices is not in general unique, and the formation of antiferromagnetic domains may occur. The appearance of more than two branches in the antiferromagnetic resonance spectrum implies the existence of a multidomain structure. The probable orientations of domains, corresponding to local minima in the free energy, have been calculated for the case of a magnetic field applied along the $[11\bar{2}]$ direction. Measurements were made on a single-crystal sample of $RbMnF_3$ at 4.2°K. For small values of applied field (<2000 Oe), the mode spectrum in the X-band region consisted of six branches. There is good agreement between the computed frequencies for the domains as a function of applied field and the experimentally observed values. The agreement implies that domain rotation takes place. A pronounced hysteresis in the resonance absorption, measured as a function of applied field, for certain resonances has been observed. The hysteresis has been attributed to domain-wall motion.

INTRODUCTION

In this paper we report the indirect observation of antiferromagnetic domains in a single-crystal sample of $RbMnF_3$. With a magnetizing field applied in the $[11\bar{2}]$ direction, the fact that the antiferromagnetic resonance (AFMR) mode spectrum contained more than two branches confirmed the existence of domains. Comparison of the experimental points with theoretical calculations implies that the majority of the domain orientations are distributed among the three directions which correspond to local minima in the free energy. Since these directions change with applied field it follows that domain rotation takes place. We have also observed a strong hysteresis in the resonance absorption which appears to be due to a redistribution of the domain pattern through domain-wall motion.

Experimental studies[1] have shown that $RbMnF_3$ is a simple cubic antiferromagnet ($T_N=82.6$°K), with the Mn^{2+} ions forming a two-sublattice antiparallel configuration. The combination of large exchange ($H_{ex}=8.9\times10^5$ Oe) and small crystalline anisotropy ($H_A=4K/3M\approx4$ Oe) permit AFMR to be observed at X-band frequencies without the need for very large magnetic fields. AFMR in applied fields large enough to cause the magnetization to be flopped has been observed by Freiser *et al.*[2] Ince[3] and Cole and Ince[4] have reported AFMR data for the low-field regime, where the flopped configuration is not in general achieved. For a single-domain model one expects the AFMR spectrum to consist of two branches, one of which becomes field-independent in the flopped state. However, in the previous work additional resonances were observed which could be attributed to a multidomain structure. The population of local minima in the energy surface takes place as a result of stresses in the crystal.[5] The latter are induced either during crystal growth or during the process of cooling to low temperatures. By domain we mean a region of the crystal in which the periodic antiferromagnetic arrangement of spins is undisturbed.

Néel[6] predicted that the formation of domains would occur in antiferromagnets which have more than one preferred direction of magnetization. However, since antiferromagnets possess little or no net magnetic moment there is no tendency for domains to form in order to reduce the dipolar energy. Thus, antiferromagnetic domain boundaries are probably ill defined and are greatly influenced by local imperfections and internal strains. However, the thickness of a domain wall is probably the same order of magnitude as for ferromagnets, since it is determined by the exchange and anisotropy energy.

Li[7] has discussed the conditions which allow the formation of antiferromagnetic domains. He has shown from thermodynamic considerations that, despite the lack of dipolar energy, domain formation may still be favored if the entropy term in the free energy is dominant. A small crystalline anisotropy, such as is found in $RbMnF_3$, is conducive to domain formation.[7] Lattice imperfections help to stabilize the domain boundaries.

$RbMnF_3$ retains its cubic structure down to very low temperatures (<4°K) with no measurable distortion. Thus, the formation of T domains, as discussed by Roth[8] in connection with NiO, is not expected to occur. On cooling a single crystal of $RbMnF_3$ in zero magnetic

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field through the antiferromagnetic ordering temperature domains will form, the magnetization within a given domain being aligned along one of the four easy axes, which are the ⟨111⟩ directions. According to Roth's classification[8] these domains will be of the S type. Within the boundary, or wall, which separates domains of different orientations a gradual rotation of the antiferromagnetic spin vectors takes place. The distribution of domains among the easy axes is most probably random, and will be greatly influenced by local strains.

SUBLATTICE ORIENTATIONS

In the case of an applied magnetic field the equilibrium direction of the antiferromagnetic arrangement within a given domain, defined as the orientation of the vector $\mathbf{M}=\frac{1}{2}(\mathbf{M}_1-\mathbf{M}_2)$, where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations, is found by minimizing the total free energy or, alternatively, by equating the total torque on $\mathbf{M}_1$ and $\mathbf{M}_2$ to zero. In the experiments to be described, the magnetizing field was applied in the $[11\bar{2}]$ direction. We shall therefore consider the problem of determining the orientation of the magnetic sublattices when the applied field is in the (110) plane. The applied field $\mathbf{H}$, and $\mathbf{M}_1$ and $\mathbf{M}_2$ are not necessarily coplanar. It is assumed that the temperature is low enough for the assumption $|\mathbf{M}_1|=|\mathbf{M}_2|=|\mathbf{M}|$ to be valid.

The total free energy $\mathcal{H}$ is equal to the sum of Zeeman, anisotropy, and exchange energies. For simplicity, the magnetostrictive term will be ignored.

The free energy can be written as

$$\begin{aligned}\mathcal{H}/\mu_0 M=&-H\{\cos\alpha\,(\cos\theta_1+\cos\theta_2)\\&+\sin\alpha\,[\sin\theta_1\cos(\phi_1-\beta)+\sin\theta_2\cos(\phi_2-\beta)]\}\\&-\tfrac{3}{16}H_A\{\sin^4\theta_1\sin^2 2\phi_1+\sin^4\theta_2\sin^2 2\phi_2\\&+\sin^2 2\theta_1+\sin^2 2\theta_2\}+H_{ex}\{\sin\theta_1\sin\theta_2\cos(\phi_1-\phi_2)\\&+\cos\theta_1\cos\theta_2\},\end{aligned}\tag{1}$$

where (θ_1, ϕ_1), (θ_2, ϕ_2), and (α, β) are the spherical coordinate angles of $\mathbf{M}_1$, and $\mathbf{M}_2$, and $\mathbf{H}$, respectively. The angles θ_1, θ_2, and α are measured from the [001] direction, while ϕ_1, ϕ_2, and β are referred to the [100] axis. The spherical angles of $\mathbf{M}$ are (θ, ϕ) and are defined by the equations

$$\begin{aligned}\theta_1&=\theta+t_\theta,\\ \theta_2&=\pi-(\theta-t_\theta),\\ \phi_1-\beta&=\phi-t_\phi,\\ \phi_2-\beta&=\pi+(\phi+t_\phi).\end{aligned}\tag{2}$$

The small angles t_θ and t_ϕ account for the tilting of $\mathbf{M}_1$ and $\mathbf{M}_2$ into the direction of the applied field.

By forming the partial derivatives of $\mathcal{H}$ with respect to the spherical angles of $\mathbf{M}_1$ and $\mathbf{M}_2$, keeping α and β fixed, and equating the expressions to zero, a set of four simultaneous equations is obtained. When the applied field is constrained to lie in the [110] plane, i.e., $\beta=\frac{1}{4}\pi$, we find

$$t_\theta\approx(H/2H_{ex})\,(\sin\alpha\cos\theta\cos\phi-\cos\alpha\sin\theta)\tag{3}$$

and

$$t_\phi\approx(H/2H_{ex})\,(\sin\alpha\sin\phi/\sin\theta).\tag{4}$$

With some algebra, and making use of the additional Eqs. (3) and (4), the following fourth-order transcendental equation is obtained:

$$\begin{aligned}&\cos^4\phi+\cos^3\phi[(\cot\alpha-0.5\tan\alpha)\cot\theta]-\cos^2\phi/2\sin^2\theta\\&+\tfrac{1}{2}\cos\phi\{[(1+3\cos 2\theta)/(4\cot\alpha\sin^2\theta\tan\theta)]\\&-\cot\theta\cot\alpha\}+[(1+3\cos 2\theta)\cot^2\theta/\sin^2\theta]=0.\end{aligned}\tag{5}$$

The roots of Eq. (5) may be found in a straightforward manner using computational methods. Imaginary roots or values for which $|\cos\phi|>1$ are discarded. The remaining roots, together with the appropriate values of α and θ, are substituted back into the starting set of simultaneous equations in order to derive the values of H, t_θ, and t_ϕ. Finally, the physically allowable solutions are tested in order to determine whether they correspond to maxima or minima in the free energy.

When $\mathbf{M}_1$, $\mathbf{M}_2$, and $\mathbf{H}$ lie in the (110) plane the partial derivatives $\partial\mathcal{H}/\partial\phi_1$ and $\partial\mathcal{H}/\partial\phi_2$ vanish, and Eq. (5) no longer holds. This special case has been discussed elsewhere,[4] and the transcendental equation is

$$(-4H^2/3H_{ex}H_A)\sin 2(\theta-\alpha)=\sin 2\theta(1+3\cos 2\theta).\tag{6}$$

Consider the application of a magnetizing field in the $[11\bar{2}]$ direction. It is found that there are four stable equilibrium directions of $\mathbf{M}$. Two of these, which we denote as I and II, lie in the same plane as the applied field. In configuration I, $\mathbf{M}$ lies along [111] when H is zero. As the field strength is increased, $\mathbf{M}$ remains flopped along [111], the sublattice vectors canting into the direction of the field by a small angle of approximate magnitude $H/2H_{ex}$. For configuration II, the magnetization initially points along $[11\bar{1}]$. Increasing the magnitude of the field from zero causes $\mathbf{M}$ to rotate gradually towards the [111] direction. Assuming H_A to be 4.08 Oe, it is found that the equilibrium becomes unstable when H is approximately equal to 1600 Oe.

The remaining two equilibrium directions, denoted as III and IV, are energetically equivalent. They correspond to the initial direction of $\mathbf{M}$ being $[\bar{1}11]$ and $[\bar{1}1\bar{1}]$, respectively. As H is increased, $\mathbf{M}$ rotates gradually towards $[\bar{1}10]$ and attains the flopped position for a field of about 3300 Oe. Orientation I has the lowest energy, while II corresponds to the highest energy because the angle between $\mathbf{M}$ and $\mathbf{H}$ is small. Orientations III and IV have intermediate energy. The relative configuration energies are illustrated in Fig. 1. Note that the constant energy term $\mu_0 M H_{ex}$ has been subtracted out.

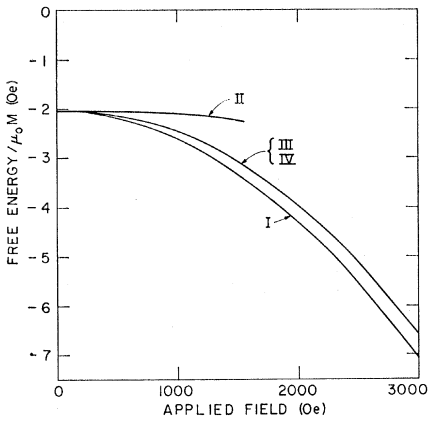

FIG. 1. Relative energy of equilibrium configuration versus applied field. The constant energy term $\mu_0 MH_{ex}$ has been subtracted out.

EXPERIMENTS

AFMR experiments were performed with a single-crystal 5-mm cube of $RbMnF_3$. The specimen was mounted on the end wall of a TE_{10n} multimode X-band cavity, which was immersed in liquid helium at 4.2°K. The sample could be rotated about a [110] axis. Resonances were observed using dc detection with magnetic field sweep and pen recorder display. The AFMR spectrum with the magnetizing field in the $[11\bar{2}]$ direction was found to consist of three pairs of mode branches, which could be associated with four different domain orientations. The resonances which corresponded to orientation I, which is the absolute minimum energy direction, exhibited the strongest absorption. The resonance absorptions for the remaining three orientations were much weaker and showed strong dependence on the magnetizing history of the sample. Very pronounced hysteresis in the intensity of these latter resonances was induced by cycling the applied field over the range 0–1000 Oe.

The experimentally observed resonances are compared with the theoretical mode curves in Fig. 2. A sketch of the four domain orientations in zero applied field is shown also. The procedure for computing the resonant frequencies as a function of applied field has been described in Ref. 4. The value of H_A, 4.08 Oe for this particular sample, was obtained from a best fit between the computed and experimental data. The resonances for the equivalent directions III and IV were not detectable for fields in excess of 600 Oe. For direction II, the resonances were barely discernible and were not detected at fields greater than about 400 Oe.

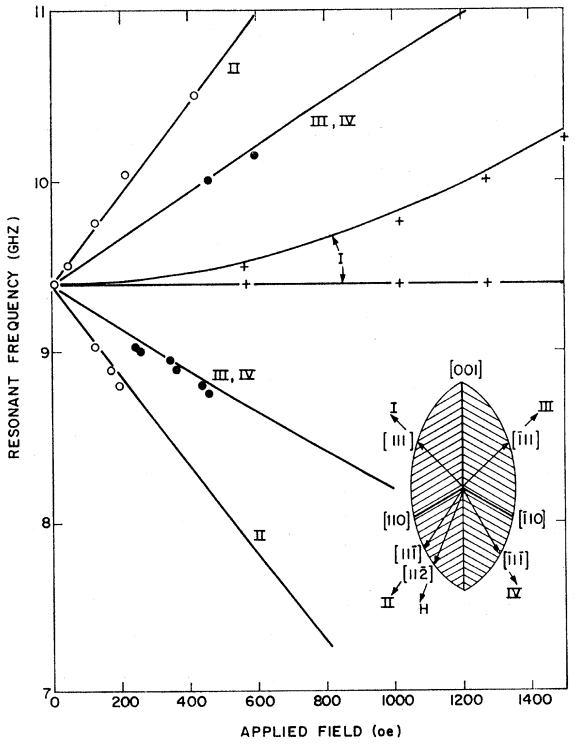

FIG. 2. Comparison of computed and experimental AFMR frequencies as a function of applied field. For the theoretical calculations H_A was assumed to be 4.08 Oe. The domain orientations for small applied field are also shown.

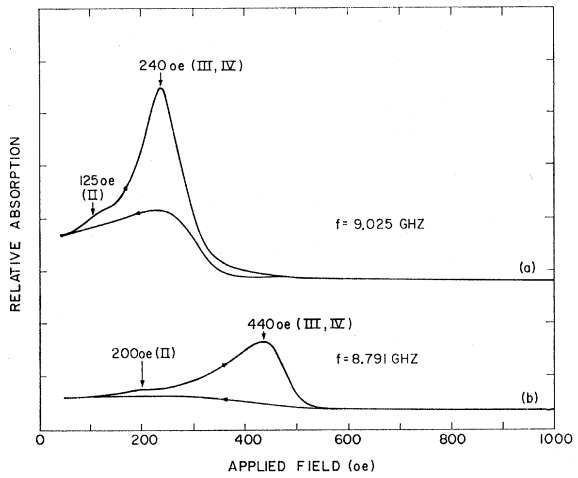

FIG. 3. Relative absorption versus applied field. The arrows indicate the direction in which the applied field was varied.

Figure 3(a) illustrates the hysteresis in the resonance absorption observed when the applied field was increased from zero to 1000 Oe and then reduced again to zero. The resonances shown correspond to equilibrium directions II, III, and IV. The frequency of 9.025 GHz was chosen to avoid absorption from the wings of the resonances corresponding to equilibrium direction I, which would have masked the much weaker resonances. Referring to the curve for increasing field strength, the larger of the two resonances shown had an asymmetric line shape, while the weaker resonance was just visible. Further, the general absorption was largest in the region

of zero applied field. On decreasing the magnetizing field from its maximum value the asymmetry in the line shape of the larger resonance was more pronounced than for increasing field, and the absorption peak was reduced. Further, the very weak resonance had disappeared. The minimum cycling time was limited to a few seconds by the response time of the electromagnet. The hysteresis in the absorption did not noticeably differ for a cycle time of about half an hour.

When the frequency was decreased, the resonances for the domains along directions II, III, and IV moved to higher field values, the absorption peak became smaller and the hysteresis in the absorption intensities was more pronounced, as illustrated in Fig. 3(b). The diminution in absorption implies that the net volume of domains II, III, and IV shrinks, while the domains corresponding to the flopped state, I, grow. The frequency in this example was 8.791 GHz. A peak was observed at 440 Oe when the field strength was increased. On reversing the field it was found that the absorption peak had almost vanished.

Figure 4 illustrates the effect of varying the range of the magnetic field sweep at constant frequency. Traces *b* through *g* were obtained by sweeping the field from zero to a maximum value, $\leq$1000 Oe, before lowering the recorder pen onto the paper. For clarity the curves have been offset from each other. It is seen that the domain resonance peak at 440 Oe remained prominent in the direction of decreasing field if the upper sweep limit was restricted to about 550 Oe. In order to obtain traces *k* through *m* the magnetic field was swept from zero to 1000 Oe, then reduced to some minimum value greater than zero with the recorder pen raised. Finally, with the pen lowered the field was increased to 1000 Oe. It is seen that the full absorption intensity of the resonance peak at 440 Oe was not recovered unless the field was reduced almost to zero. In trace *m* the weak resonance at 200 Oe is still not detectable. It is important to note that no additional resonances appeared, nor were the original resonances shifted to higher field values. Hence although domain rotation does, of course, occur under the action of the applied field, it does not appear to be the origin of the hysteresis. Figure 5 shows a similar set of curves taken at a frequency of 8.893 GHz.

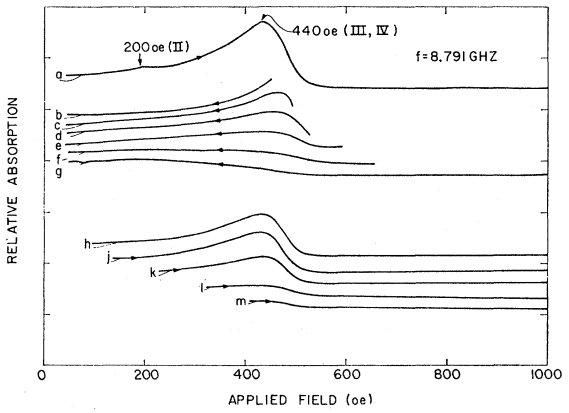

FIG. 4. Relative absorption versus applied field (f=8.791 GHz). In order to obtain traces *b* through *g*, the field was swept from zero to its maximum value before lowering the recorder pen onto the paper. Traces *h* through *m* were obtained by varying the field from 0 to 1000 Oe and then reduced to some minimum before lowering the pen. Arrows indicate the direction of field variation.

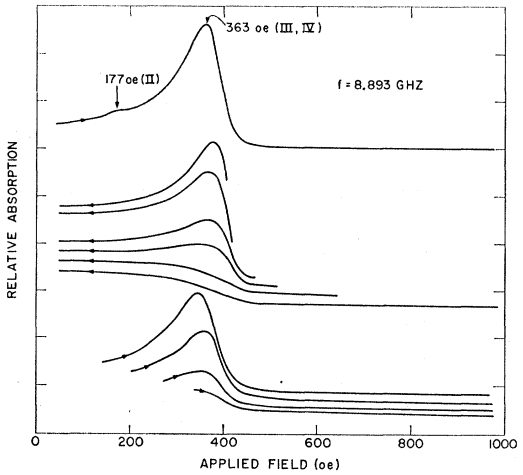

FIG. 5. Relative absorption versus applied field (f=8.893 GHz). Arrows indicate the direction of field variation.

CONCLUSIONS

We believe that the observed hysteresis, asymmetry of the line shape, and variation in resonance absorption intensity with frequency are due to a redistribution of antiferromagnetic domains via domain-wall motion. Regions having spin orientation I grow at the expense of the other domains. The good agreement between the calculated AFMR mode branches and experimental points implies that coherent rotation of the magnetization within individual domains also takes place.

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