Ultrasonic Attenuation near and above the Spin-Flop Transition of MnF_2

Y. SHAPIRA AND J. ZAK*

Francis Bitter National Magnet Laboratory,† Massachusetts Institute of Technology, Cambridge, Massachusetts

(Received 29 January 1968)

The ultrasonic attenuation in MnF_2 was measured at 4.2°K in steady magnetic fields up to 150 kG. These measurements were performed with 8 to 310-Mc/sec longitudinal and shear sound waves propagating along the [100], [110], and [001] crystallographic axes. The most prominent features of the data are: (1) For several acoustical modes, a sharp peak in the attenuation occurs near the spin-flop transition at 93 kG. This peak is observed only if **H** is within a few tenths of a degree from the [001] axis. (2) For two acoustical modes, a large absorption edge, as a function of magnetic field, exists near the spin-flop transition. The edge is observed only if **H** is within ~2° from the [001] axis. Supplementary antiferromagnetic-resonance experiments, at X band, were performed on the same single crystal of MnF_2. Several mechanisms which may be responsible for the observed magnetic field variation of the ultrasonic attenuation are discussed.

I. INTRODUCTION

In the last two decades there has been a number of investigations of the ultrasonic behavior of antiferromagnets. These investigations dealt for the most part with the temperature variation of the ultrasonic velocity (or Young's modulus) and attenuation (or internal friction) at zero magnetic field. Particular attention was paid to changes in the ultrasonic properties which occur at or near the Néel temperature,[1–14] or, in some substances, at a temperature where the magnetic structure changes from one antiferromagnetic phase to another.[6–8,11,15] Recently, there have been several investigations of the ultrasonic behavior of antiferromagnets near magnetic phase transitions which occur, at a fixed temperature, as the external magnetic field is varied. Thus, an absorption peak for ultrasonic waves was observed by Shapira[16] near the spin-flop transition of the two uniaxial antiferromagnets MnF_2 and $(Cr_2O_3)_{0.94}(Al_2O_3)_{0.06}$. An ultrasonic absorption peak and a change in Young's modulus near the spin-flop transition of chromium were later reported by Street *et al.*[11] The magnetic field variation of the ultrasonic velocity and attenuation in $RbMnF_3$ was recently investigated by Melcher *et al.*[13]

In the present paper we report the results of ultrasonic attenuation measurements in MnF_2, carried out at 4.2°K and in steady magnetic fields up to 150 kG. Some preliminary results of this study, for ultrasonic waves propagating along the tetragonal crystallographic axis, [001], were briefly mentioned earlier.[16] Here, we present a more complete set of results for longitudinal and shear ultrasonic waves propagating along the [100], [110], and [001] axes.

MnF_2 is antiferromagnetic below the Néel temperature $T_N=67$°K. The magnetic behavior of MnF_2 at $T<T_N$ is, in many cases, well approximated by that of an ideal uniaxial antiferromagnet composed of two interpenetrating identical sublattices.[17] This fact makes MnF_2 a favorite candidate for experimental tests of various aspects of the theory of antiferromagnetism. At zero magnetic field, the two sublattices of MnF_2 have equal and opposing magnetic moments which are directed along the tetragonal axis. If a strong magnetic field **H** is applied along the tetragonal axis, then the sublattice magnetizations rotate to a direction which is nearly perpendicular to **H**. The rotation of the sublattice magnetizations takes place at a certain magnetic field H_{sf} which depends on temperature. This sudden realignment of the sublattice magnetizations is known as the spin-flop transition.[18] For MnF_2, $H_{sf}=93$ kG at 4.2°K.[17,19] When the external magnetic field is not directed exactly along the tetragonal axis but makes a small angle θ with it, the rotation of the sublattice mag-

* Permanent address: Department of Physics, Technion-Israel Institute of Technology, Haifa, Israel.

† Supported by the U.S. Air Force Office of Scientific Research.

[1] R. Street and B. Lewis, Nature **168**, 1036 (1951); Phil. Mag. **1**, 663 (1956); R. Street and J. H. Smith, J. Phys. Radium **20**, 82 (1959).

[2] M. E. Fine, Phys. Rev. **87**, 1143 (1952).

[3] K. P. Belov, G. I. Katayev, and R. Z. Levitin, J. Appl. Phys. Suppl. **31**, 153 (1960).

[4] L. A. Yevtushchenko and R. Z. Levitin, Fiz. Metal. Metalloved. **12**, 155 (1961) [English transl.: Soviet Phys.—Metals and Metallography **12**, 139 (1961)].

[5] J. R. Neighbours, R. W. Oliver, and C. H. Stillwell, Phys. Rev. Letters **11**, 125 (1963).

[6] M. E. de Morton, Phys. Rev. Letters **10**, 208 (1963).

[7] R. Street, Phys. Rev. Letters **10**, 210 (1963).

[8] D. I. Bolef and J. de Klerk, Phys. Rev. **129**, 1063 (1963).

[9] E. J. O'Brien and J. Franklin, J. Appl. Phys. **37**, 2809 (1966).

[10] K. Tani and H. Mori, Phys. Letters **19**, 627 (1966).

[11] R. Street, B. C. Munday, B. Window, and I. Williams, J. Appl. Phys. **39**, 1050 (1968).

[12] K. Walther, Solid State Commun. **5**, 399 (1967).

[13] R. L. Melcher, D. I. Bolef, and R. W. H. Stevenson, Solid State Commun. **5**, 735 (1967).

[14] B. Golding, Phys. Rev. Letters **20**, 5 (1968).

[15] R. W. Makkay, G. H. Geiger, and M. E. Fine, J. Appl. Phys. **33**, 914 (1962).

[16] Y. Shapira, Phys. Letters **24A**, 361 (1967).

[17] S. Foner, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. I, p. 383; Phys. Rev. **107**, 683 (1957); J. Phys. Radium **20**, 21 (1959); Phys. Rev. **130**, 183 (1963).

[18] For a simple discussion of the spin-flop phenomenon see T. Nagamiya, K. Yosida, and R. Kubo, Advan. Phys. **4**, 1 (1955) or Ref. 17.

[19] I. S. Jacobs, J. Appl. Phys. Suppl. **32**, 61 (1961).

netizations takes place over a finite magnetic field interval which increases with θ, i.e., the spin-flop transition is broadened as θ increases. Neighbours *et al.*[5] studied the ultrasonic attenuation in MnF_2 near the Néel temperature and at zero (or low) magnetic field. The present high-field experiments, on the other hand, were undertaken for the purpose of studying the ultrasonic attenuation near and above the spin-flop transition of MnF_2.

A brief discussion of the experimental techniques is given in Sec. II. The results of ultrasonic attenuation measurements for several acoustical modes of propagation are presented in Sec. III. In Sec. IV the experimental results are discussed and are compared to predictions based on several theoretical models.

In addition to the ultrasonic work, a brief account is given in this paper of conventional (not acoustical) antiferromagnetic-resonance experiments, at X band, which were carried out on the same crystal of MnF_2 as used in the ultrasonic experiments. The purpose of the antiferromagnetic-resonance (AFMR) experiments was to provide supplementary information, independent of the ultrasonic measurements, about the behavior of this particular sample at fields near H_{sf}. AFMR experiments in MnF_2 were performed earlier by Foner[17] and by Johnson and Nethercot.[20]

II. EXPERIMENTAL TECHNIQUE

Ultrasonic-attenuation measurements were performed on several single crystals of MnF_2 at 4.2°K and in magnetic fields up to 150 kG. All the specimens used in the present experiments were cut from one large single crystal of MnF_2, oriented by x rays, and their end faces were lapped for ultrasonic work. The specimens were approximately cubic with an edge which varied from 6 to 15 mm. Longitudinal and shear ultrasonic waves, with frequencies from 8 to 310 Mc/sec, were generated with X-cut and Y-cut quartz transducers operating at their fundamental frequency (8–12 Mc/sec) or in one of the overtones. Acoustical bonds were made with Dow Corning 200 silicon oil having a viscosity of 25 000 centistoke at 25°C. Standard ultrasonic-pulse techniques were used. The magnetic field variation of the attenuation was measured by gating one of the acoustical echoes, integrating it, and recording the output as a function of H. The time separation between successive ultrasonic echoes was used to determine the sound velocity V_s, at $H=0$, with an estimated accuracy of $\pm 2\%$. Steady magnetic fields up to 150 kG were produced by a water-cooled Bitter-type solenoid.

As we shall see below, the magnitude of the attenuation changes near the spin-flop transition is very sensitive to the angle θ between $\mathbf{H}$ and the [001] crystallographic axis. Thus, a change of a fraction of a degree in θ can cause a drastic change in the attenuation. Since the *absolute* orientation of the sample was not known to within a fraction of a degree, the following procedure was used: The sample was mounted with its [001] axis oriented to within $\sim 1°$ of the magnetic field direction. The orientation of the sample was then adjusted until the largest change in attenuation was observed near the spin-flop transition. It was then *assumed* that the latter orientation was $\mathbf{H}||[001]$.

Conventional (not acoustical) AFMR experiments were carried out at 4.2°K using 8–12-kMc/sec radiation. The samples, which were mounted on the shorting end wall of an X-band waveguide, were immersed in liquid helium. Resonance absorption was observed as a change in the reflected power, monitored with a crystal detector, as a function of H. The magnetic field in these experiments was aligned to within $\sim 1°$ of the [001] crystallographic axis.

III. EXPERIMENTAL RESULTS

A. Ultrasonic Attenuation

The magnetic field variation of the acoustical attenuation of longitudinal and shear ultrasonic waves propagating along the [100], [110], and [001] crystallographic axes was measured at 4.2°K in magnetic fields up to 150 kG. The zero-field ultrasonic velocities at 4.2°K for the various acoustical modes which were excited in these experiments are listed in Table I. Each mode is characterized by the direction of its propagation vector $\mathbf{q}$, and the direction of ion displacement $\boldsymbol{\xi}$. The results of the attenuation measurements are given separately for each mode.

1. $\mathbf{q}||\boldsymbol{\xi}||$*[001]. Longitudinal Wave*

Attenuation measurements were carried out with 8–220-Mc/sec waves. With $\mathbf{H}||[001]$ axis, a sharp absorption peak was observed in all cases at 92.4 ± 0.5 kG. This field value is equal, within experimental error, to the value $H_{sf}=93\pm 2$ kG for the spin-flop field as determined by magnetization measurements.[19] No hysteresis was observed in the ultrasonic absorption peak. Figure 1 shows some typical recorded data.

The height $\Delta\alpha$ of the absorption peak, for $\mathbf{H}||[001]$, was large in all cases. For example, in the case of the 30-Mc/sec wave whose attenuation is shown in Fig. 1, $\Delta\alpha \gtrsim 4$ dB/cm. The height of the absorption peak decreased very rapidly as the angle θ between $\mathbf{H}$ and the [001] axis was increased from zero. As a result, the absorption peak could be observed only when θ was less than a few tenths of a degree. In many cases the absorption peak for $\mathbf{H}||[001]$ was so large that no ultrasonic echoes could be observed at the spin-flop transition. For this reason, and because of the strong dependence of $\Delta\alpha$ on θ, no accurate results concerning the height of the absorption peak or its frequency dependence can be given.

2. $\mathbf{q}||\boldsymbol{\xi}||$*[110]. Longitudinal Wave*

Measurements were performed with 30–130-Mc/sec waves. With $\mathbf{H}||[001]$ axis, a sharp absorption peak was

[20] F. M. Johnson and A. H. Nethercot, Phys. Rev. **104**, 847 (1956); **114**, 705 (1959).

TABLE I. The velocity of sound, V_s, at 4.2°K for several modes of propagation.

q	ξ	Type of mode	V_s(10^5 cm/sec)
[001]	[001]	longitudinal	6.6
[001]	in (001) plane	shear	2.84
[100]	[100]	longitudinal	5.2
[100]	[010]	shear	4.4
[100]	[001]	shear	2.84
[110]	[110]	longitudinal	6.6
[110]	[1$\bar{1}$0]	shear	1.67
[110]	[001]	shear	2.84

observed at 92.4±0.5 kG. The width of the peak was, in some samples, as narrow as 300 G. The peak could be observed only when θ did not exceed a few tenths of a degree. When the magnetic field was aligned as close as possible to the [001] axis, the height of the absorption peak for a 30-Mc/sec wave was $\Delta\alpha \gtrsim 4$ dB/cm. In some runs with 30- and 50-Mc/sec waves, a small increase in the attenuation, of about 0.1 to 0.2 dB/cm, was observed above H_{sf}. Figure 2 shows an example of these results.

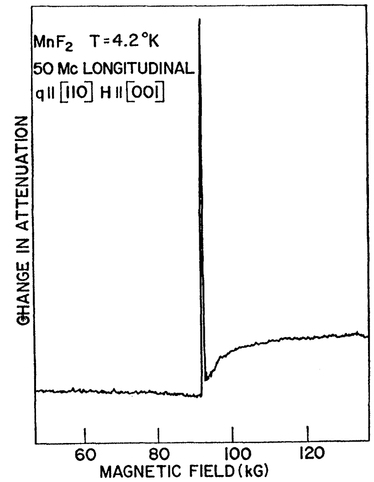

FIG. 2. Recorder tracing of the attenuation of a 50-Mc/sec longitudinal wave with **q** || [110] and **H** || [001]. The recorder response to the attenuation is nonlinear.

3. q || ξ || [*100*]. *Longitudinal Wave*

Attenuation measurements were made with 30–310-Mc/sec waves. With **H** near the [001] axis, two distinct features of the attenuation were observed. First, the attenuation increased at the spin-flop transition and remained high at $H > H_{sf}$. With **H** as close as possible to the [001] axis, the increase in attenuation was abrupt and its magnitude was so large that in most runs no acoustical echoes could be seen at $H > H_{sf}$. Figure 3 shows the results obtained with a 30-Mc/sec wave for which the attenuation increase at the spin-flop transition was greater than 10 dB/cm. When **H** was not directed exactly along [001], but rather made a small angle ($\theta \lesssim 2°$) with this axis, a gradual increase in the absorption was observed at $H > H_{sf}$, i.e., the absorption edge was broadened and moved to higher fields. For $\theta \gtrsim 2°$ the attenuation did not depend strongly on H in the range 0–150 kG.

A second feature, which was observed only when the angle θ did not exceed a few tenths of a degree, was a sharp spike in the ultrasonic attenuation which occurred near the spin-flop transition. This spike was not distinct when **H** was exactly along the [001] axis because it was masked by the absorption edge (see Fig. 3). However, for an angle θ of the order of a few tenths of a degree, the absorption edge moved to slightly higher fields and the spike could be seen clearly. This is illustrated in Fig. 4., which shows a recorder tracing of the attenuation of a 190-Mc/sec wave for **H** near the [001] axis. In this trace the attenuation at $H > H_{sf}$ passed through a broad maximum at fields just above H_{sf} and then started to

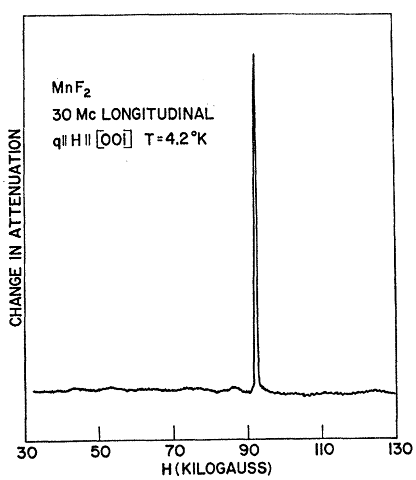

FIG. 1. Recorder tracing of the attenuation of a 30-Mc/sec longitudinal wave with **q** || **H** || [001].

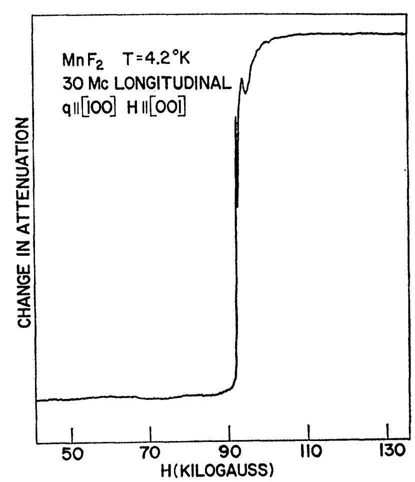

FIG. 3. Recorder tracing of the attenuation of a 30-Mc/sec longitudinal wave with **q** || [100] and **H** || [001]. The recorder response to the attenuation is nonlinear. The straight line at fields well above H_{sf} corresponds to the absence of any detectable acoustical echoes.

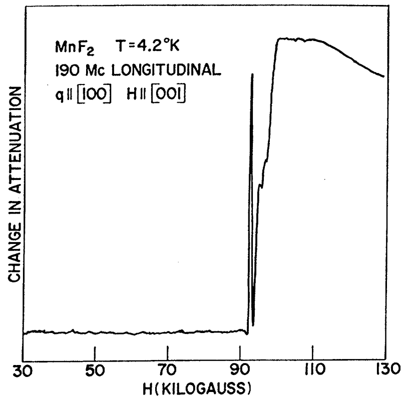

FIG. 4. Recorder tracing of the attenuation of a 190-Mc/sec longitudinal wave with **q** ∥ [100] and **H** near the [001] axis. The recorder response to the attenuation is nonlinear.

decrease with increasing **H**. Similar traces were obtained in the same experimental run with 270- and 310-Mc/sec waves.

4. **q**∥*[001]*, *ξ in (001) Plane. Shear Wave*

Measurements were made with 8–230-Mc/sec waves. The most prominent feature in the attenuation-versus-H curve was a large sharp spike in the attenuation which occurred at 92.4±0.5 kG, i.e., near the spin-flop transition. As in the cases described earlier, the height of the spike was very sensitive to the angle θ. In addition to the spike, an increase in the absorption at fields above H_{sf} was usually observed when the angle between **H** and [001] was small. This increase in absorption was very sensitive to the angle θ and disappeared at $\theta \gtrsim 2°$. In most of the experiments the orientation of the vector ξ in the (001) plane was not controlled. However, in two experimental runs ξ was oriented along the [100] and along the [110] axes. The results in these cases were similar to those obtained when the direction of ξ was not controlled.

5. **q**∥*[110]*, *ξ*∥*[001]*. *Shear Wave*

Measurements were carried out with 10-, 30-, and 50-Mc/sec waves. In one run a very small increase (~0.1 dB/cm) in the attenuation of a 30-Mc/sec wave was observed near the spin-flop transition when **H** was near the [001] axis. This small increase might have been fortuitous. In two other runs with 30-Mc/sec waves no change in attenuation was observed at H_{sf} to an accuracy of ~0.1 dB/cm. A single run was performed with 10- and 50-Mc/sec waves. No change in the attenuation greater than ~0.3 dB/cm was observed with these two frequencies in the field interval 0–140 kG.

6. **q**∥*[110]*, *ξ*∥*[1̄10]*. *Shear Wave*

Measurements were made with 10–230-Mc/sec waves. With **H** directed along the [001] axis, the absorption increased abruptly at H_{sf} and no acoustical echoes could be seen at $H>H_{sf}$. For 30 and 50-Mc/sec waves the increase of the attenuation at the spin-flop transition was larger than 20 dB/cm. Figure 5 shows some of the results obtained with a 50-Mc/sec wave.

The dependence of the absorption edge on the angle θ between **H** and the [001] axis was measured. As θ was increased from zero, the absorption edge moved to higher fields and the increase in the absorption took place over a wider field interval. In order to observe the absorption edge in fields up to 150 kG it was necessary for θ to be less than ~1.5°. For θ very close to zero a sharp peak in the absorption was sometimes observed at H_{sf}, superposed on the absorption edge. It is noteworthy that the results for shear waves with **q**∥[110], ξ∥[1̄10] resemble those for longitudinal waves with **q**∥[100].

7. **q**∥*[100]*, *ξ*∥*[001]*. *Shear Wave*

Attenuation measurements were carried out with 30- and 50-Mc/sec waves. For H∥[001] the attenuation increased slightly at the spin-flop transition and remained at the new (higher) level at $H>H_{sf}$. The magnitude of this absorption edge for 50-Mc/sec waves was ~0.2 dB/cm.

8. **q**∥*[100]*, *ξ*∥*[010]*. *Shear Wave*

Measurements were made with 30- and 50-Mc/sec waves. With **H**∥[001], a small absorption edge was observed near the spin-flop transition. The magnitude of the absorption edge was ~0.3 dB/cm for 30-Mc/sec waves. The edge was not observed for angles $\theta \gtrsim 2°$.

B. Antiferromagnetic Resonance

AFMR experiments were carried out at X-band frequencies, at 4.2°K, and with the magnetic field aligned

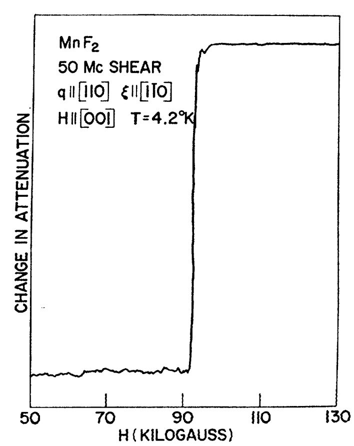

FIG. 5. Recorder tracing of the attenuation of a 50-Mc/sec shear wave with **q** ∥ [110], ξ ∥ [1̄10], and **H** ∥ [001]. The recorder response to the attenuation is nonlinear. The straight line at fields well above H_{sf} corresponds to the absence of any detectable acoustical echoes.

to within $\sim 1°$ of the [001] axis. The aim of these experiments was twofold. First, to determine the field H_1 at which $\omega=0$ for the low-frequency spin mode[17] when $\mathbf{H}\|[001]$. Second, to obtain an estimate of the width of the AFMR line in our sample.

The field H_1 is very close to the spin-flop transition field H_{sf} (see Sec. IV A1 below). It can be obtained from the field $H_r(\omega)$ at which AFMR occurs for the frequency ω by using the relation[17]

$$\omega = g\mu_B[H_1 - H_r(\omega)], \tag{1}$$

where μ_B is the Bohr magneton and g is the g factor. The g factor for MnF_2, in the antiferromagnetic state and when $\mathbf{H}\|[001]$, is known only to an accuracy of several percent.[20] However, in the present experiments ω is sufficiently small that no significant error in H_1 will arise if we let $g=2.0$.

Two experimental runs with two different samples were performed. In the first run the sample was about 6 mm thick and the frequency was 9.36 kMc/sec. A resonance was observed at 90.3 ± 0.3 kG, which gives $H_1=93.6\pm0.3$ kG. The full width at half-height of the resonance was about 500 G. In the second run a thinner sample, about 1 mm thick, was used. From the resonance fields for the frequencies 8.36, 10.75, and 11.64 kMc/sec we obtained $H_1=93.3\pm0.3$ kG. The full width at half-height of the resonance was about 100 G. Thus, the values of H_1 for both samples were in good agreement with each other although the linewidths differed considerably. The latter difference is probably related to the difference in the thickness of the two samples.

In deriving H_1 from the values of $H_r(\omega)$ we assumed that in the AFMR experiments the magnetic field was exactly parallel to the [001] axis. However, since $H_r(\omega)$ increases as the angle θ increases from zero,[17] and since in practice θ is never exactly zero, the above values for H_1 are systematically higher than the true value of H_1. It is estimated that this systematic error did not exceed 1% in the present experiments. Our results for H_1 are in good agreement with the value $H_1=93.4$ kG extrapolated from the data of Johnson and Nethercot[20] assuming, as these authors did, that $g=2.00$.

IV. DISCUSSION

The ultrasonic results presented in the preceding section indicate the existence of two, apparently distinct, phenomena. First there is a sharp absorption peak which occurs at, or near, the spin-flop transition when $\mathbf{H}$ is within a few tenths of a degree of the [001] axis. Second, an absorption edge for ultrasonic waves exists at the spin-flop transition when $\mathbf{H}\|[001]$. The magnitude of either the absorption peak or the absorption edge varies considerably from one ultrasonic mode (characterized by $\mathbf{q}$ and $\boldsymbol{\xi}$) to another. For some modes the absorption peak and/or absorption edge either do not exist or are very small, whereas for other modes the magnitude of either or both of these phenomena is so large that no acoustical echoes are seen. In the following paragraphs several physical mechanisms which may be responsible for the experimental results are discussed.

A. Absorption Peak near the Spin-Flop Transition

We first consider mechanisms which may be responsible for the sharp absorption peak which occurs at, or near, the spin-flop transition.

1. *Acoustical Antiferromagnetic Resonance at $H \lesssim H_{sf}$*

The existence of an absorption peak near the spin-flop transition was predicted by Peletminskii[21] and by Savchenko.[22] These authors discussed the interaction of acoustical phonons with low-frequency magnons in a uniaxial antiferromagnet. The magnetoacoustic coupling at $H<H_{sf}$ is due to the modulation of the anisotropy energy by the elastic strain. According to Peletminskii and Savchenko this coupling should lead to a strong interaction between the acoustical mode and the low-frequency magnon mode when the frequencies and wave vectors of the two modes coincide. The process which gives rise to this strong interaction is one in which a single phonon is annihilated and a single magnon is created. One may view this process as an "acoustical antiferromagnetic resonance" (hereafter AAFMR). In order to conserve energy and momentum in such a process, the frequencies and wave vectors of the phonon must coincide with those of the magnon. An analogous situation in ferromagnets was considered earlier by Kittel.[23]

The dispersion relation for the low-frequency magnon mode of a uniaxial antiferromagnet, at $T=0°$K and when $\mathbf{H}$ is directed along the preferred axis, is[24]

$$\omega = \gamma(2H_EH_A + H_A^2 + bH_E^2d^2q^2)^{1/2} - \gamma H, \tag{2}$$

where ω is the frequency, γ is the gyromagnetic ratio, H_E is the exchange field, H_A is the anisotropy field, q is the magnon wave number, d is the lattice constant, and b is a constant of order unity. The appropriate values for MnF_2 are[17] $H_E=5.6\times10^5$ G, $H_A=7.8\times10^3$ G, and $d\sim4\times10^{-8}$ cm. Note that $H_E\gg H_A$. AAFMR for a sound wave with frequency ω should occur at the field $H_R(\omega)$, which is given by

$$H_R(\omega) = [2H_EH_A + H_A^2 + bH_E^2d^2(\omega^2/V_s^2)]^{1/2} - (\omega/\gamma), \tag{3}$$

where V_s is the sound velocity. The resonance field $H_r(\omega)$ for conventional AFMR (with microwaves) can be obtained from Eq. (3) by deleting the third term in the brackets, on the right-hand side of this equation. In

[21] S. V. Peletminskii, Zh. Eksperim. i Teor. Fiz. **37**, 452 (1959) [English transl.: Soviet Phys.—JETP **10**, 321 (1960)].

[22] M. A. Savchenko, Fiz. Tverd. Tela **6**, 864 (1964) [English transl.: Soviet Phys.—Solid State **6**, 666 (1964)].

[23] C. Kittel, Phys. Rev. **110**, 836 (1958).

[24] F. Keffer, H. Kaplan, and Y. Yafet, Am. J. Phys. **21**, 250 (1953).

the present ultrasonic experiments $\omega \lesssim 2\times 10^9$ rad/sec, which is a very small frequency compared to $\gamma(2H_EH_A)^{1/2}$. It is easy to show that in this case $H_R(\omega)$ is close to the spin-flop transition field H_{sf}, which is given approximately by[17]

$$H_{sf}\cong(2H_EH_A)^{1/2}. \quad (4)$$

From the theory of conventional AFMR[17] one expects, by analogy, that AAFMR may be observed only when **H** makes a very small angle with the [001] axis. Physically, the frequency of the low-frequency spin mode does not go to zero at any field unless θ is zero. Instead, for any finite θ there exists a minimum value ω_{min} for the frequency of this spin mode in the range $H\leq H_{sf}$. In order to observe acoustical (or ordinary) AFMR at a given frequency, it is necessary that ω not be lower than ω_{min} by an amount exceeding the reciprocal of the lifetime of the magnon, i.e., energy must be conserved within the uncertainty in the energy of the magnon. In the present case the uncertainty in the energy of the magnon, estimated from the AFMR linewidth of several hundred gauss, is larger than the sound frequency. Using this fact and charts which are given in Ref. 17 it was estimated that AAFMR with rf sound waves can be observed only if θ is less than a few tenths of a degree.

Although the theory for AAFMR seems to account for the observed ultrasonic absorption peak near H_{sf}, and the extreme sensitivity of this peak to the angle θ, there are reasons to doubt the validity of this explanation. According to the theory,[21,22] AAFMR at $H<H_{sf}$ should not be observed with longitudinal waves propagating along the [001] axis.[25] This conclusion disagrees with the experimental results (see Fig. 1).

Another difficulty with the AAFMR explanation arises when one examines the spin-flop transition in detail. An approximate expression for the spin-flop field H_{sf} was given above [Eq. (4)]. More careful considerations of the spin-flop phenomenon[26,27] indicate, however, that there are three physically important magnetic fields $H_1>H_3>H_2$. First there is the field H_1, which is given by

$$H_1=(2H_EH_A+H_A^2)^{1/2}. \quad (5)$$

At this field the frequency of the spin mode, which is given by Eq. (2), vanishes for $q=0$. H_1 represents an upper limit for the field at which the spins can be oriented, at equilibrium, along (and opposite to) the [001] axis. A second field H_2, which is given by

$$H_2=(2H_EH_A+H_A^2)^{1/2}(2H_E-H_A)/(2H_E+H_A) \quad (6)$$

represents a lower limit for the field at which the spins at equilibrium can be nearly normal to the [001] axis. A third field H_3, which is given by

$$H_3=(2H_EH_A-H_A^2)^{1/2} \quad (7)$$

represents the field at which the free energies of the two spin configurations (parallel and nearly normal to the [001] axis) are equal to each other. In an increasing magnetic field, an *ideal* sample of MnF_2 should undergo a spin-flop transition at H_1, whereas in a decreasing field the transition should occur at the lower field H_2. Thus, the spin-flop transition in an ideal sample should show a hysteresis. A *real* sample of MnF_2 may undergo a spin-flop transition at H_3 both for an increasing and a decreasing magnetic field, i.e., it may not show "supercooling" and "superheating" effects. For MnF_2 the field difference H_1-H_2 is 1.3 kG, approximately, and H_3 is the geometric mean of H_1 and H_2. The field $H_R(\omega)$ at which AAFMR should occur for $\omega\lesssim 10^9$ rad/sec satisfies the relations $H_1>H_R(\omega)>H_3>H_2$. There are now two possibilities. First, there may be a hysteresis in the spin-flop transition. In this case AAFMR should be observed only in an increasing magnetic field, but not in a decreasing field. Second, the spin-flop transition may occur at H_3 for both increasing and decreasing fields. The AAFMR should not be observed then in either an increasing or a decreasing magnetic field. Either conclusion disagrees with observation.

The preceding argument is weakened when one takes into account the finite linewidth of the AAFMR. If the AAFMR linewidth is comparable to or larger than $H_R(\omega)-H_3$, and if the sample undergoes a transition at H_3 for both an increasing and a decreasing magnetic field, then AAFMR may be observed near H_3. In the present experiments $H_R(\omega)-H_3\cong 600$ G. The AAFMR linewidth is estimated, from the ordinary AFMR linewidth at 8–12 Gc, to be equal to 100 G to within an order of magnitude. Thus, the AAFMR linewidth may be comparable to $H_R(\omega)-H_3$.

In principle one should be able to determine H_1 exactly from experiment and then one can calculate $H_R(\omega)$, H_2, and H_3. Thus one should be able to find out at which of these fields does the ultrasonic absorption peak occur. In the present experiments, however, the experimental uncertainties in the value of H_1, and in the value at which the ultrasonic absorption peak occurs, are comparable to the small differences between H_1, H_2, H_3, and $H_R(\omega)$. Thus, one cannot say with certainty at which of these fields the ultrasonic peak occurs.

In order to ascertain whether the absorption peak is due to a resonance phenomenon one must be able to distinguish between effects due to the resonance and effects due to the phase transition. Had the frequency ω of the ultrasonic wave been in the microwave range, the resonance field $H_R(\omega)$ would have been several kG lower than H_1, H_2, and H_3, and this question could be settled. On the basis of the present data on rf ultrasonic waves, however, this question cannot, in the opinion

[25] A similar situation exists for the magnetoacoustic resonance in ferromagnets (see Ref. 23).

[26] F. Keffer, in *Handbuch der Physik*, edited by S. Flügge and H. P. J. Wign (Springer-Verlag, Berlin, 1966), Vol. XVIII/2, p. 1.

[27] A. I. Akhiezer, V. G. Bar'yakhtar, and M. I. Kaganov, Usp. Fiz. Nauk **71**, 533 (1960) [English transl.: Soviet Phys.—Usp. **3**, 567 (1961)].

of the authors, be answered with certainty. [*Note added in proof*. The theories of Peletminskii and Savchenko concentrate on a process in which a phonon is absorbed and a magnon is created. Near the spin-flop transition. however, higher-order processes, which involve more than one magnon, may become important. This was recently considered by K. Tani, Phys. Letters **26A**, 419 (1968)].

2. *Domains near H_{sf}*

With the magnetic field directed along the [001] axis, the orientation of the spins changes abruptly at H_{sf}. At $H<H_{sf}$ the spins are oriented along [001], whereas at $H>H_{sf}$ the spins are nearly perpendicular to [001]. At $H=H_{sf}$ the orientation of the spins relative to the [001] axis is not uniquely determined. It is then possible for the sample to break into domains with different spin directions.[28] If an applied elastic stress favors a particular spin orientation, then it will cause domains having this spin orientation to grow at the expense of others. Changes in the relative size of the domains may be accomplished by a displacement of domain walls. The alternating elastic stress which is associated with the ultrasonic wave may then lead to a periodic motion of the domain walls which will result in an increase in the ultrasonic absorption. Ultrasonic absorption due to motion of domain walls in antiferromagnets has been repeatedly suggested in the literature (see, for example, Ref. 1).

B. Absorption at $H>H_{sf}$

With **H** directed along the [001] axis, an absorption edge is observed near H_{sf} for certain ultrasonic modes of propagation. This effect is particularly large for longitudinal waves propagating along the [100] axis, and for shear waves with $\mathbf{q}\|[110]$ and $\boldsymbol{\xi}\|[1\bar{1}0]$. In the following paragraphs we shall consider several mechanisms which may be responsible for the ultrasonic absorption at $H>H_{sf}$.

1. *Spin Waves at $H>H_{sf}$*

In a uniaxial antiferromagnet, with **H** directed along its preferred axis, there exist two spin modes at $H>H_2$. Unless H is just above H_2, one of these spin modes has a very high frequency (compared to radio frequencies) for all values of q. This so-called spin-flop mode can interact with microwaves,[17] but is not expected to interact strongly with rf sound waves except, possibly, near H_2. The second spin mode has the following dispersion relation for small wave numbers[26,27]:

$$\omega=V(H)q, \tag{8}$$

where $V(H)$ varies with H according to the relation

$$V(H)=V_0[1-(H/2H_E)^2]^{1/2}. \tag{9}$$

The velocity V_0 depends on the direction of $\mathbf{q}$ but is independent of the magnitude of q. Since $\omega=0$ for $q=0$, the spin mode described by Eq. (8) cannot be excited with microwaves as long as **H** is parallel to the preferred axis. This mode is sometimes referred to as the "nonactivated" mode.

The interaction of the nonactivated spin waves with ultrasonic waves was investigated theoretically by Bar'yakhtar *et al.*[29] According to these authors, longitudinal sound waves couple strongly to the nonactivated spin waves because of the modulation of the exchange interaction by the strains associated with the sound wave. Bar'yakhtar *et al.* considered the process in which one phonon is annihilated and one magnon is created. They predicted that a magnetoacoustic resonance will occur when the sound velocity V_s is equal to the spin-wave velocity $V(H)$. This condition follows from the conservation of energy and momentum. Since neither V_s nor $V(H)$ depend on the magnitude of q, the condition $V_s=V(H)$ is fulfilled simultaneously for all values of q. If the magnetic field **H** is tilted away from the preferred axis, then Eq. (8) is no longer valid. Instead, the frequency ω for a wave with $q=0$ becomes finite, i.e., there exists an energy gap. For sufficiently large θ this energy gap is large enough to exclude the possibility of a resonance absorption of rf phonons by the nonactivated spin mode. Thus, strong magnetoacoustic coupling may be observed only when **H** is near the preferred axis.

In order to compare the experimental results with the theory of Bar'yakhtar *et al.*, it is necessary to know the velocity $V(H)$ of the nonactivated spin waves. In the present experiments $H/2H_E<0.14$, so that the difference between $V(H)$ and V_0 is less than 1%. We shall therefore neglect the field dependence of $V(H)$ and let $V(H)=V_0$. The velocity V_0 is calculated in the Appendix. The result is

$$V_0=(8S/\hbar)\{-J^A[J_c^Fc^2\alpha_3^2+J_a^Fa^2(\alpha_1^2+\alpha_2^2)-J^A(a^2\alpha_1^2+a^2\alpha_2^2+c^2\alpha_3^2)]\}^{1/2}, \tag{10}$$

where J^A is the antiferromagnetic exchange constant between the Mn ion at (0, 0, 0) and the one at $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$, J_a^F is the ferromagnetic exchange constant between the Mn atom at (0, 0, 0) and the one at (1, 0, 0), J_c^F is the ferromagnetic exchange constant between the Mn atom at (0, 0, 0) and the one at (0, 0, 1), S is the spin of the Mn ion, α_i are the direction cosines of $\mathbf{q}$ in the [100], [010], and [001] coordinate system, and a and c are the lattice constants of MnF_2. Letting[30] $J^A=-1.76$°K, $J_c^F=0.32$°K, $J_a^F<0.05$°K, and[26] $a=4.87$ A, $c=3.31$ A, $S=\frac{5}{2}$, one obtains the following values: $V_0=2.3\times10^5$ cm/sec for $\mathbf{q}\|[100]$ or $\mathbf{q}\|[110]$, and $V_0=1.7\times10^5$ cm/sec for $\mathbf{q}\|[001]$.

Comparison of the calculated values of V_0 with the

[28] For a review of domains in antiferromagnets see M. M. Farztdinov, Usp. Fiz. Nauk, **84**, 611 (1964) [English transl.: Soviet Phys.—Usp. **7**, 855 (1965)].

[29] V. G. Bar'yahktar, M. A. Savchenko, and V. V. Tarasenko, Zh. Eksperim. i Teor. Fiz. **49**, 944 (1965) [English transl.: Soviet Phys.—JETP **22**, 657 (1966)].

[30] A. Okazaki, K. C. Turberfield, and R. W. H. Stevenson, Phys. Letters **8**, 9 (1964).

velocities of the various acoustical modes (Table I) indicates that V_0 and V_s are of the same order of magnitude. However, the velocities of the acoustical modes which are strongly attenuated at $H>H_{sf}$ (the mode with $\mathbf{q}\|\xi\|[100]$, and the one with $\mathbf{q}\|[110]$, $\xi\|[1\bar{1}0]$) are not equal to V_0. This fact by itself does not invalidate the supposition that the large attenuation at $H>H_{sf}$ is due to a resonance absorption of phonons by the nonactivated spin mode. As mentioned earlier, the condition $V_s=V(H)$ follows from the conservation of energy and momentum in a process where a phonon is annihilated and a magnon is created. However, if one takes into account the finite lifetime τ of the magnon, one arrives at the conclusion that $V(H)$ need not be exactly equal to V_s. Assuming $\tau\lesssim 10^{-9}$ sec, which does not seem to be unreasonable, one can explain the discrepancy between V_s and $V(H)$ which exists in the present experiments with rf sound waves.

Whereas the preceding argument may account for the discrepancy between $V(H)$ and the velocities of those acoustical modes which are strongly attenuated at $H>H_{sf}$, it does not explain why other modes are not strongly attenuated at $H>H_{sf}$. The dramatic difference in the attenuation of the various acoustical modes at $H>H_{sf}$ is not related in any obvious way to a match between the velocity of the spin wave and that of the sound wave.[31]

According to the theory of Bar'yakhtar *et al.* shear waves are not coupled to the nonactivated spin waves at $H>H_{sf}$. This selection rule, which arises from the assumption of isotropic exchange interaction, should apply also to the shear mode $\mathbf{q}\|[110]$, $\xi\|[1\bar{1}0]$ in MnF_2. The experimental fact that this shear mode is strongly attenuated at $H>H_{sf}$ is not explained, therefore, by the theory of Bar'yakhtar *et al.*

Three final remarks about the phonon-magnon interaction at $H>H_{sf}$: (a) Bar'yakhtar *et al.* considered a process in which one phonon is annihilated and one magnon is created. A complete theory must take into account other, more complicated, processes or show that they are unimportant. (b) Recently, Akhiezer and Borovik[32] considered nonlinear spin waves in antiferromagnets. These waves may also interact with sound waves at $H>H_{sf}$. (c) The vast difference between the attenuation of the $\mathbf{q}\|\xi\|[100]$ mode and that of the $\mathbf{q}\|\xi\|[110]$ mode, as well as the difference between the attenuation of the $\mathbf{q}\|[110]$, $\xi\|[1\bar{1}0]$ mode and that of the $\mathbf{q}\|[100]$, $\xi\|[010]$ mode, cannot be explained by any theory which approximates MnF_2 by a uniaxial antiferromagnet which has a cylindrical symmetry. Some form of an anisotropy in the (001) plane must be introduced.

2. Domains at $H>H_{sf}$

At fields above the spin-flop transition and with $\mathbf{H}$ directed along the preferred axis, the equilibrium orientation of the spins of an ideal uniaxial antiferromagnet is not uniquely determined by free-energy considerations. This is because the free energy of the uniaxial antiferromagnet is not changed when the magnetizations of both sublattices are rotated simultaneously about the preferred axis by an arbitrary angle. As a consequence, there exists the possibility that the antiferromagnet will break into domains with different spin orientations. The existence of domains may then give rise to ultrasonic absorption.

The domain picture which was presented in the previous paragraph, in which it was assumed that MnF_2 can be approximated by an ideal uniaxial antiferromagnet which has a cylindrical symmetry, fails to explain the vast difference between the attenuation of: (a) the ultrasonic mode with $\mathbf{q}\|\xi\|[100]$ and that of the mode $\mathbf{q}\|\xi\|[110]$, and (b) the ultrasonic mode with $\mathbf{q}\|[110]$, $\xi\|[1\bar{1}0]$ and that of the mode $\mathbf{q}\|[100]$, $\xi\|[010]$. The origin of the difficulty is the assumption that MnF_2 possesses a cylindrical symmetry about the [001] axis. We therefore assume instead that there is some anisotropy in the basal plane of MnF_2, i.e., there exists a set of preferred directions for the spins in the basal plane. Since this set of directions must have the symmetry of MnF_2, there are several spin configurations for which the free energy is minimum. Thus, there exists the possibility of a domain structure. If an elastic stress is applied, then the free-energy densities of two domains with different spin orientations may change by the same amount for both domains, or they may change by different amounts. In the former case there is no reason why one domain should grow at the expense of the other so that domain-wall motion, and the resultant ultrasonic attenuation, are not expected. In the opposite case, ultrasonic attenuation due to domain-wall motion is expected.

Two models for domains at $H>H_{sf}$ were considered. In the first model it was assumed that: (a) The anisotropy energy of a spin with a given orientation is the same for both sublattices, and (b) the $\langle 100\rangle$ equivalent directions are preferred energetically. In this case there are domains with spins along and opposite to the [100] direction[33] and those with spins along and opposite to the [010] direction. These domains have no net magnetic moment in the basal plane. A symmetry operation which transforms a [100] domain into a [010] domain (and vice versa) is a reflection in the (110) plane. This symmetry exists for the unstrained lattice. If this symmetry remains also in the strained lattice, then the free-energy densities of the [100] and [010] domains remain equal to each other.

Assuming that the anisotropy energy depends only

[31] It is interesting that among the longitudinal modes the one with the lowest velocity is the one which is most strongly attenuated at $H>H_{sf}$ and that the same is true for the shear modes. It is not clear whether this fact is of any particular significance.

[32] I. A. Akhiezer and A. E. Borovik, Zh. Eksperim. i Teor. Fiz. **52**, 508, 1332 (1967) [English transl.: Soviet Phys.—JETP **25**, 332, 885 (1967)].

[33] More accurately, the spins are in the (010) plane making a small angle with the [100] axis.

on the relative orientation of the spins with respect to the crystal, one can show that certain strains change the free-energy densities of the [100] and [010] domains by the same amount. In particular, one can show that this is the case for strains associated with the following acoustical modes: (1) longitudinal $\mathbf{q}||\xi||[001]$, (2) longitudinal $\mathbf{q}||\xi||[110]$, (3) shear $\mathbf{q}||[100]$, $\xi||[010]$, (4) shear $\mathbf{q}||[110]$, $\xi||[001]$, and (5) shear $\mathbf{q}||[001]$, $\xi||[110]$. The strains associated with all of these five modes leave the (110) plane as a mirror plane. Experimentally, all the five modes, with the possible exception of the last one, are not strongly attenuated at $H>H_{sf}$. Thus, the fact that modes No. 2 and 3 are not strongly attenuated at $H>H_{sf}$, whereas the modes $\mathbf{q}||\xi||[100]$ and $\mathbf{q}||[110]$ $\xi||[1\bar{1}0]$ are strongly attenuated, is consistent with this domain model. It is noteworthy that if the $\langle 110\rangle$ equivalent directions were the preferred directions for the spins, then the attenuation (due to domains) of sound waves with $\mathbf{q}||\xi||[100]$ should have been small at $H>H_{sf}$, which disagrees with observation.

Another model for the domains which was considered is one in which the anisotropy in the basal plane is due to Moriya's single-ion anisotropy mechanism.[26,34] As shown by Moriya,[34] such an interaction may lead to domains which have a net magnetic moment (Dzyaloshinsky moment) in the basal plane. This net moment can be directed along any of the four equivalent $\langle 100\rangle$ directions.[33] Symmetry considerations indicate that the free-energy density of all the domains should change by the same amount when strains are produced by the following acoustical modes: (1) longitudinal $\mathbf{q}||\xi||[001]$, (2) longitudinal $\mathbf{q}||\xi||[110]$, and (3) shear $\mathbf{q}||[100]$, $\xi||[010]$. These "selection rules" for attenuation due to domain-wall motion are consistent with the observation that none of these acoustical modes is strongly attenuated at $H>H_{sf}$.

The change in the spin orientation at the spin-flop transition of MnF_2 has some similarity to the change in the spin orientation near the Morin transition of α-Fe_2O_3. In both cases the spins change their orientation from a direction along the preferred axis to a direction (or directions) which are nearly perpendicular to this axis. The basic difference is that the spin-flop transition is often observed at constant temperature and in a changing magnetic field, whereas the Morin transition is observed at zero field by varying the temperature. It is interesting that at temperatures above the Morin transition of α-Fe_2O_3, the internal friction is higher than at temperatures below this transition. Makkay *et al.*,[15] who observed this effect, attributed it to the existence of domains at temperatures above the Morin transition, where the spins are in the basal plane. In the case of α-Fe_2O_3 it is known from the work of Dzyaloshinsky[35] and Moriya[36] that there exists a set of preferred directions for the spins in the basal plane.

[34] T. Moriya, Phys. Rev. **117**, 635 (1960).
[35] I. Dzyaloshinsky, J. Phys. Chem. Solids **4**, 241 (1958).
[36] T. Moriya, Phys. Rev. **120**, 91 (1960).

The preceding discussion indicates that the assumption that domains, with spins along certain preferred axes, exist at $H>H_{sf}$ leads to a reasonably successful explanation of the ultrasonic attenuation of the various acoustical modes. There remains, however, the difficulty of explaining the origin of the anisotropy energy in the basal plane. The second model for the domains was based on Moriya's single-ion anisotropy mechanism. Using the known parameters[37] for this interaction in MnF_2, we expect it to be very weak, which casts doubt on the validity of the second model for the domains. Also, we cannot give any *a priori* reason why the $\langle 100\rangle$ directions should be preferred, as required by the first model for the domains. Therefore, until more tangible evidence for the existence of domains in the spin-flop state of MnF_2 is available, the explanation of the ultrasonic data in terms of domains cannot, in our view, be regarded as an entirely convincing explanation.

V. CONCLUSIONS

The most prominent features of the ultrasonic attenuation measurements are: (1) the sharp peak in the attenuation coefficient near the spin-flop transition, and (2) an absorption edge, as a function of H, which occurs near the spin-flop transition. The magnitude of either of these phenomena depends on the particular ultrasonic mode of propagation and is either zero or very small for some modes. Both phenomena can be seen only when $\mathbf{H}$ is near the [001] axis.

Several physical mechanisms which may be responsible for the experimental data have been considered. It was shown that existing theories which concentrate on processes in which a single phonon is annihilated and a single magnon is created do not give a satisfactory explanation of the data. In particular, selection rules against the interaction of longitudinal waves (at $H<H_{sf}$) and shear waves (at $H>H_{sf}$), which are derived from these theories, disagree with observation. Some models for ultrasonic absorption due to domain-wall motion are consistent with the data, but, in the opinion of the authors are not entirely convincing at present. It therefore appears that further investigation of the poorly explored spin-flop state in MnF_2, and other antiferromagnets, is necessary for a satisfactory explanation of the ultrasonic data.

ACKNOWLEDGMENTS

We wish to thank our colleagues S. Foner and A. Misetich for many useful discussions. We also wish to thank M. Kelly, E. J. Alexander, and W. Tice for technical assistance.

APPENDIX

The spin-wave spectrum in the spin-flop state of MnF_2 was derived using the following model. The anti-

[37] R. R. Sharma, T. P. Das, and R. Orbach, Phys. Rev. **149**, 257 (1966).

ferromagnetic exchange interaction between the Mn ion at (0, 0, 0) and the one at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ was assumed to be equal to $-2J^A\mathbf{S}_1\cdot\mathbf{S}_2$, where $\mathbf{S}_1$ and $\mathbf{S}_2$ are the spins of these ions. The weaker ferromagnetic interactions between the Mn ion at (0, 0, 0) and the ions at (1, 0, 0) and (0,0,1) were assumed to be given by similar terms with J_a^F and J_c^F, respectively. All exchange interactions except those between the first-, second-, and third-nearest neighbors were neglected. The various exchange constants are given in Ref. 30. The magnetic field was assumed to be along the [001] axis. The anisotropy energy for each spin was assumed to be of the form $-K\cos^2\varphi$, where K is a constant and φ is the angle between the direction of the spin and $\mathbf{H}$.

The spin-wave spectrum in the spin-flop state was derived both classically and quantum mechanically. The results of both derivations are identical. Also, by letting $J_a^F=J_c^F=0$, our results reduce to those given by Keffer.[26] Some details of the classical derivation are given below. The quantum-mechanical derivation was carried out using a procedure similar to the one employed by Kanamori and Yosida[38] in their treatment of a uniaxial antiferromagnet, with $\mathbf{H}$ normal to the preferred axis.

Let $\mathbf{S}_i^A$ be the spin of an ion which is at the ith site of the A sublattice (and similarly for $\mathbf{S}_j^B$) and let J_{ij}^{AB} be the exchange constant between the spin on the ith site of the A sublattice and the one on the jth site of the B sublattice (with a similar definition for J_{ik}^{AA}). The classical equations of motion for the spins are

$$\hbar(d\mathbf{S}_i^A/dt)=g\mu_B\mathbf{S}_i^A\times(\mathbf{H}+\mathbf{H}')+\mathbf{S}_i^A \times 2\Big(\sum_j J_{ij}^{AB}\mathbf{S}_j^B+\sum_k J_{ik}^{AA}\mathbf{S}_k^A\Big),$$

$$\hbar(d\mathbf{S}_j^B/dt)=g\mu_B\mathbf{S}_j^B\times(\mathbf{H}+\mathbf{H}')+\mathbf{S}_j^B \times 2\Big(\sum_k J_{jk}^{BA}\mathbf{S}_k^A+\sum_l J_{jl}^{BB}\mathbf{S}_l^B\Big), \quad \text{(A1)}$$

where $\mathbf{H}'$ is directed along $\mathbf{H}$ and has a magnitude which is related to the anisotropy field by the relation $H'=H_A\cos\varphi$. The indices i and k in Eqs. (A1) are for the A sublattice, while j and l are for the B sublattice. In the spin-flop state the equilibrium orientations of the magnetizations of the two sublattices are not collinear. In this case it is convenient to express the spins in Eqs. (A1) in the following coordinate systems: Spins in the A sublattice are expressed in the coordinate system x', y', z', with the z' axis along the equilibrium position of $\mathbf{S}_i^A$. All spins in the B sublattice are expressed in the coordinate system x'', y'', z'', with the z'' axis along the equilibrium position of $\mathbf{S}_j^B$. The angle φ_0 between the equilibrium position of $\mathbf{S}_i^A$ (or $\mathbf{S}_j^B$) and $\mathbf{H}$ can be easily found to be[26]

$$\cos\varphi_0=H/(2H_E-H_A), \quad \text{(A2)}$$

where

$$H_E=(2S/g\mu_B)\sum_j J_{ij}^{AB} \quad \text{(A3)}$$

is the antiferromagnetic exchange field. By going through the regular procedure[26] for solving Eqs. (A1) one finds the following expression for the spin-wave spectrum in the spin-flop state:

$$(\hbar\omega)^2=\alpha\beta+4S^2J(\mathbf{q})\cos2\varphi_0\pm2SJ(\mathbf{q})[\alpha\cos2\varphi_0+\beta], \quad \text{(A4)}$$

where

$$\alpha=g\mu_B \times(H\cos\varphi_0+H_A\cos^2\varphi_0-H_E\cos2\varphi_0+H_E^F)-2SJ^F(\mathbf{q}),$$

$$\beta=g\mu_B \times(H\cos\varphi_0-H_A\sin^2\varphi_0-H_E\cos2\varphi_0+H_E^F)-2SJ^F(\mathbf{q}), \quad \text{(A5)}$$

$$H_E^F=\frac{2S}{g\mu_B}\sum_k J_{ik}^{AA} \quad \text{(A6)}$$

is the ferromagnetic exchange field, and

$$J(\mathbf{q})=\sum J^{AB}(\mathbf{R}^{AB})\exp(i\mathbf{q}\cdot\mathbf{R}^{AB}), \quad \text{(A7)}$$

$$J^F(\mathbf{q})=\sum J^{AA}(\mathbf{R}^{AA})\exp(iq\cdot\mathbf{R}^{AA}). \quad \text{(A8)}$$

In Eq. (A7) the summation is over distances $\mathbf{R}^{AB}$ between spins on different sublattices, while in (A8) the summation is on distances $\mathbf{R}^{AA}$ between spins in the same sublattice. By taking into account only the exchange interactions between first-, second-, and third-nearest neighbors, and after assuming that $H\ll H_E$, $H_A\ll H_E$, one obtains the following final expressions for the spin-wave spectrum in the spin-flop state:

$$(\hbar\omega_1)^2=64J^AS^2\{J^A[(q_xa)^2+(q_ya)^2+(q_zc)^2] -J_c^F(q_zc)^2-J_a^F[(q_xa)^2+(q_ya)^2]\}, \quad \text{(A9)}$$

$$(\hbar\omega_2)^2=(g\mu_B)^2(H^2-H_0^2)+64J^AS^2 \times\{J^A[(q_xa)^2+(q_ya)^2+(q_zc)^2] -J_c^F(q_zc)^2-J_a^F[(q_xa)^2+(q_ya)^2]\}, \quad \text{(A10)}$$

where a and c are the lattice constants of MnF_2, $H_0^2=2H_EH_A$, and the x, y, and z axes are along [100], [010], [001], respectively. Equation (A9) corresponds to the nonactivated mode, whereas Eq. (A10) corresponds to the spin-flop mode. Equation (A9) leads to Eq. (10) in the text.

[38] J. Kanamori and K. Yosida, Progr. Theoret. Phys. (Kyoto) **14**, 423 (1955).