Ultrasonic Behavior near the Spin-Flop Transition of Cr_2O_3

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The attenuation of longitudinal and shear ultrasonic waves exhibits a peak at the spin-flop transition of Cr_2O_3. This peak is attributed to the presence of antiferromagnetic domains near the transition. The attenuation peak for longitudinal waves is accompanied by a dip in the phase velocity. The temperature variation of the spin-flop-transition field H_{sf} was measured from 4.2 to 305°K and is in very good agreement with earlier differential magnetization data obtained by Foner and Hou. At 4.2°K, $H_{sf}=60.3\pm1.0$ kG, whereas at 305°K, $H_{sf}=121.2\pm1.5$ kG.

I. INTRODUCTION

In the last decade there have been a number of investigations of the magnetic phase transitions of antiferromagnets.[1] One such phase transition is the spin-flop transition which occurs in uniaxial antiferromagnets of the easy-axis type. In such an antiferromagnet, the application of a sufficiently strong magnetic field **H** along the easy axis causes the sublattice magnetizations to rotate from directions parallel to this axis to directions in the plane which is normal to this axis. If **H** is exactly along the easy axis, then the reorientation of the spins is abrupt and is accompanied by a discontinuous change in the magnetic moment of the sample. This phenomenon is called a spin-flop transition, and it occurs at a magnetic field H_{sf} which is a function of temperature.[2]

Ultrasonic propagation near the spin-flop transition of several antiferromagnets was studied in the last two years.[3–6] These studies show that the transition is accompanied by large changes in the ultrasonic attenuation coefficient and velocity. Such "anomalies" in the ultrasonic properties, aside from their intrinsic interest, may serve as a convenient means of observing the spin-flop transition and studying the temperature variation of H_{sf}. Previously,[3] we have observed a peak in the ultrasonic attenuation near the spin-flop transition of $(Cr_2O_3)_{0.94}(Al_2O_3)_{0.06}$ at 77°K. A study of pure Cr_2O_3 was prevented at that time by the unavailability of a sufficiently large pure single crystal. In the present work on pure Cr_2O_3 the spin-flop transition was studied ultrasonically from 4.2 to 305°K. This work is similar in many ways to our earlier work[4,6] on MnF_2 and α-Fe_2O_3.

Cr_2O_3 has a rhombohedral (trigonal) structure and is antiferromagnetic below the Néel temperature $T_N=308$°K. In the antiferromagnetic state the sublattice magnetizations, at zero applied magnetic field, point along the c axis (trigonal axis). The spin-flop transition in Cr_2O_3 was studied by Foner and Hou,[7] who measured the differential magnetization in pulsed fields. The transition field H_{sf} was found to increase from 59 kG at 4.2°K to 113 kG at 285°K. The temperature variation of H_{sf} can also be deduced from resonance and susceptibility measurements,[8] or from data on the magnetoelectric effect.[9]

II. EXPERIMENTAL TECHNIQUE

Ultrasonic measurements were carried out on a single crystal of Cr_2O_3 using pulse techniques. The single crystal was kindly provided by A. N. Mariano of the Kennecott Copper Corp. and was prepared for ultrasonic work. The sample which was used in the ultrasonic experiments had two parallel and flat c faces separated by 5 mm from each other. Longitudinal and shear waves were generated with X-cut and Y-cut quartz transducers, respectively. Acoustical bonds between the transducers and the sample were made with several bonding materials. At low temperatures either Dow Corning 200 Silicone fluid, having a viscosity of 30 000 cS at 25°C, or Nonaq stopcock grease was used. At temperatures above 250°K, Canada balsam was used. Measurements of the magnetic field dependence of the attenuation were made by gating one of the acoustical echoes, integrating it, and recording the output as a function of H. Occasionally the attenuation was measured point by point by comparing the heights of successive acoustical echoes. Measurements of the magnetic field variation of the phase velocity were carried out using a technique which has been described earlier.[10]

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[1] See, for example, V. A. Schmidt and S. A. Friedberg, J. Appl. Phys. **38**, 5319 (1967). A schematic of the phase diagram of a simple antiferromagnet is shown in Fig. 1 of this reference.

[2] For a simple discussion of the spin-flop phenomenon, see T. Nagamiya, K. Yosida, and R. Kubo, Advan. Phys. **4**, 1 (1955), or S. Foner, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. I, p. 383.

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Experiments at 4.2 and 77°K were performed in liquid helium and liquid nitrogen, respectively. Measurements at $77<T\lesssim300$°K were made in the following way. The sample was mounted on a copper block and was placed in a Dewar filled with liquid nitrogen. The liquid nitrogen was then siphoned out and the sample was allowed to warm slowly to room temperature. Typically, it took a few hours for the sample to reach room temperature. Data were taken while the temperature was changing. The temperature was measured with a platinum thermometer embedded in the copper block on which the sample was mounted. Measurements at $300\lesssim T<310$°K were carried out by first warming the sample, and the copper block on which it was mounted, with a heat gun and then placing it inside an empty Dewar. Data were taken as the temperature drifted down slowly.

High dc magnetic fields, up to 150 kG, were generated in Bitter-type solenoids at the Francis Bitter National Magnet Laboratory, M.I.T. The magnetic field in these magnets was known to an accuracy of 1%.

III. RESULTS

Measurements of the ultrasonic attenuation as a function of H were carried out at $4.2<T<310$°K and in fields up to 150 kG. Most runs were made at 77°K but two runs covered the entire temperature range from 4.2 to 310°K. The results at 77°K will be discussed first.

The attenuation of 30- and 50-MHz longitudinal waves propagating along the c axis was measured at 77°K as a function of H. With the magnetic field along the c axis a peak in the attenuation was observed at 62.3 ± 1.0 kG, which is where the spin-flop transition occurs.[7] At the field where the attenuation was maximum, no acoustical echoes could be observed. The width of the attenuation peak was about 6 kG. Figure 1 shows the attenuation peak for a 50-MHz wave.

The dependence of the attenuation peak on the angle θ between **H** and the c axis was measured with a 50-MHz longitudinal wave at 77°K and over the angular interval $0°\leqslant\theta\leqslant21°$. As θ increased, the height of the attenuation peak decreased, the peak became broader,

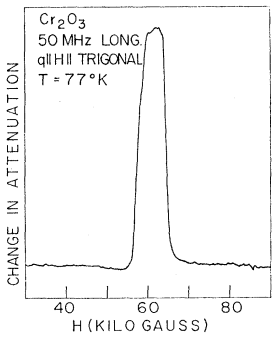

FIG. 1. Recorder tracing of the attenuation of a 50-MHz longitudinal wave at 77°K. The direction of sound propagation and **H** are both parallel to the c axis. The recorder response to to the attenuation is nonlinear.

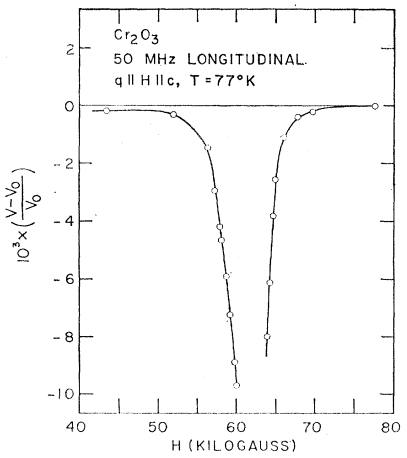

FIG. 2. Magnetic field variation of the phase velocity V of a 50-MHz longitudinal wave at 77°K. The direction of sound propagation and **H** are both parallel to the c axis.

but the field at the center of the peak did not change appreciably. At $\theta=3°$ the height of the attenuation peak decreased to approximately half its value at $\theta=0$, and the width of the peak was about 10 kG. A very small and very broad attenuation peak was still present at $\theta=21°$.

The attenuation peak for longitudinal waves was accompanied by a dip in the phase velocity V. Figure 2 shows the magnetic field variation of V for the longitudinal wave whose attenuation is shown in Fig. 1. The zero-field velocity of this wave is $V_0=(8.2\pm0.1)\times10^5$ cm/sec. As can be seen from Fig. 2, the dip in the phase velocity had a magnitude greater than 1%. In another run with a 50-MHz wave the phase velocity had a dip of more than 1.8% at H_{sf}. The exact magnitude of the dip in the phase velocity could not be measured because no acoustical echoes were observed when the attenuation was maximum.

Experiments with 11- and 25-MHz shear waves propagating along the c axis were also performed at 77°K. An attenuation peak, similar to the one for longitudinal waves, was observed at the spin-flop transition. With an 11-MHz wave, the attenuation peak at $\theta=0$ was greater than 5 dB/cm.

To determine the temperature variation of the spin-flop field, the attenuation of a 50-MHz longitudinal wave propagating along the c axis was measured as a function of the magnetic field, applied along the same axis, over the temperature interval 4.2 to 310°K. A large peak in the attenuation marked the position of the spin-flop transition. As the temperature increased, the height of the attenuation peak decreased and the peak became narrower. At 300°K the full width at half-height of the peak was about 3 kG compared with ~6 kG at 77°K. The temperature variation of H_{sf} is shown in Fig. 3. At 4.2°K, $H_{sf}=60.3\pm1.0$ kG, and at 305°K (the highest temperature at which spin flop was

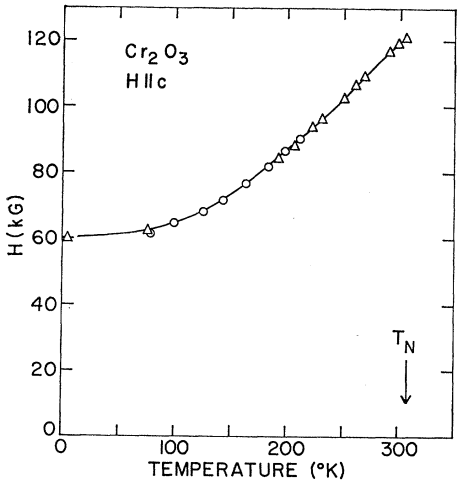

FIG. 3. Temperature variation of the spin-flop field H_{sf}.

observed) $H_{sf}=121.2\pm1.5$ kG. The spin-flop transition disappeared between 305 and 307.5°K. The temperature where this transition disappears is the temperature of the triple point, which should be slightly lower than the Néel temperature (see Ref. 1, Fig. 1).

IV. DISCUSSION

An ultrasonic attenuation peak at the spin-flop transition of antiferromagnets was discovered earlier in MnF_2, α-Fe_2O_3, $(Cr_2O_3)_{0.94}(Al_2O_3)_{0.06}$, and chromium.[3–6] The observation of an attenuation peak near the spin-flop transition of Cr_2O_3 is therefore in line with the results in other antiferromagnets.

Compared with the attenuation peak in MnF_2, the attenuation peak in Cr_2O_3 is broad and its magnitude decreases more slowly with increasing θ. The width of the spin-flop transition in the Cr_2O_3 sample measured by Foner and Hou[7] was less than 1 kG. This fact suggests that the comparatively broad ultrasonic attenuation peak observed in the present work is due to a variation in the transition field from one part of the sample to another. A dependence of the width of the spin-flop transition on the quality of the sample was observed in other antiferromagnets.[6]

The attenuation peak at H_{sf} is probably due to the motion of domain walls. Since this mechanism has already been described in detail elsewhere,[6] we shall discuss it here only briefly.

At the spin-flop transition, the orientation of the spins changes from directions parallel to the c axis to directions perpendicular to it. Assuming that the transition in different parts of the sample occurs at slightly different fields, one is led to the conclusion that at the average transition field H_{sf} some regions of the sample have their spins parallel to the c axis while other regions have their spins perpendicular to it. Thus, at H_{sf} the sample consists of several antiferromagnetic domains. If an elastic stress is then applied, it will change the free energies of domains with different spin configurations by different amounts, except in special cases. As a result, domains with lower free energy will tendto grow at the expense of domains with higher free energy. The changes in the relative sizes of domains are accomplished by displacements of domain walls. The periodic stress associated with the sound wave leads therefore to a periodic motion of domain walls. Assuming that there is some damping of the domain-wall motion, one expects the sound wave, which drives the domain walls, to be attenuated. It is well known[11] that a similar mechanism leads to ultrasonic attenuation in ferromagnets.

It should be noted that the present results for the shear waves are somewhat different from the results for shear waves in MnF_2 and α-Fe_2O_3. In MnF_2 and α-Fe_2O_3 the shear attenuation coefficient, for **H** along the c axis, exhibits a peak at H_{sf} but is also substantially higher at $H>H_{sf}$ than at $H<H_{sf}$. It is believed that in these materials the attenuation at $H>H_{sf}$ is caused by domains in which the spins point along different directions in the c plane.[4,6] In Cr_2O_3, however, the attenuation for 11- and 25-MHz shear waves is substantially the same at $H<H_{sf}$ and at $H>H_{sf}$. Apparently, the attenuation due to domain-wall motion at $H>H_{sf}$ is small in this case.

Turning to the temperature variation of H_{sf}, we note that our results are in very good agreement with those obtained by Foner and Hou[7] from differential magnetization measurements. A further comparison to Foner's resonance and susceptibility results[8] can be made by using the relation[2]

$$H_{sf}=[2H_EH_A/(1-\alpha)]^{1/2}, \quad (1)$$

where H_E is the exchange field, H_A is the anisotropy field, and $\alpha=\chi_{\parallel}/\chi_{\perp}$ is the ratio of the susceptibilities parallel and perpendicular to the c axis. Values of H_{sf} calculated from Foner's results and Eq. (1) are in very good agreement with the present data.

In conclusion, we note that ultrasonic measurements are a simple and convenient means of observing the spin-flop transition and studying the temperature dependence of the transition field.

ACKNOWLEDGMENTS

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[11] W. P. Mason, *Physical Acoustics and the Properties of Solids* (D. Van Nostrand, Inc., Princeton, N. J., 1958), Chap. 8.