AIP Journal of Applied Physics

Anomalies in the Compressional and Shear Properties of Hematite in the Region of the Morin Transition

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Citation: Journal of Applied Physics 41, 1414 (1970); doi: 10.1063/1.1658968
View online: http://dx.doi.org/10.1063/1.1658968
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/41/3?ver=pdfcov
Published by the AIP Publishing

enon is observed as shown in Fig. 2. Both c_{44}^* and c_{33}^* show rather sharp minima. The linear coupling theory outlined above is in qualitative agreement with the behavior of c_{44}^*. However, because of the very strong orientation dependence no quantitative comparisons were possible. Misorientations of less than one degree were sufficient to change qualitatively the behavior of c_{44}^* in this region, often producing a maximum in c_{44}^* on the high-field side of the minimum shown in Fig. 2. On the other hand, the c_{33}^* behavior cannot be explained on the basis of the linear theory and showed no qualitative change for small misorientations although the magnitude of the effect was very orientation sensitive.

One possible explanation, proposed by Tani,[5] is that the volume magnetostrictive interaction causes the anomalous behavior of c_{33}^* at the spin–flop transition. (This second-order effect can be described in terms of the creation or annihilation of an acoustic phonon accompanied by the scattering of a thermal magnon from one state to another.) At low fields and low temperatures, such that the spin-wave energy gap is much greater than kT, this process is inhibited. However, for $H_0 \simeq H_c$, the energy gap for the lower branch of the spin-wave spectrum approaches zero for zero wavevector, causing an enhancement of the effect. Symmetry requires that the volume magnetostriction provides coupling only to longitudinal elastic modes. Shapira and Zak[6] have observed sharp peaks in the ultrasonic attenuation for all longitudinal modes and at least one transverse mode near the spin–flop transition.

For $H_0 > H_c$ the results are complicated by the possible existence of domains and sample shape effects, and will not be discussed here.

The author is indebted to Professor D. F. Holcomb for a critical reading of the manuscript.

* Work supported by the Advanced Research Projects Agency through the Materials Science Center at Cornell University, MSC Report #1260.

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JOURNAL OF APPLIED PHYSICS VOLUME 41, NUMBER 3 1 MARCH 1970

Anomalies in the Compressional and Shear Properties of Hematite in the Region of the Morin Transition*

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Compressional (v_p) and shear (v_s) wave velocities have been measured in a hot-pressed (ρ= 5.254 g/cc) polycrystalline hematite (α-Fe_2O_3) as a function of temperature from 200° to 300°K using the pulse superposition method of McSkimin. Both v_p and v_s exhibit anomalous behavior in the region of the Morin transition ($T_M \cong 253$°K), but the effect is much more pronounced for v_s than for v_p; v_s at T_M is changed by less than 0.02% by performing the experiment in an applied field of 650 Oe. In order to test whether the observed anomalies in v_p and v_s have their origin in domain wall–stress interaction, v_p was measured along the trigonal (c) axis of a high-purity single crystal kindly lent by Y. Shapira. A sharp increase ($\Delta v_p = 0.2\%$) was observed as the crystal was cooled through T_M. In the absence of an applied field, T_M increases by 3.6°K/kbar; this acoustic determination of ($\partial T_M/\partial P$) is in agreement with that determined from neutron diffraction and NMR studies but differs from that measured by Kawai and Ôno. The v_p jump at T_M did not vanish when the experiment was repeated with a field of 650 Oe applied in the c plane; this is in marked contrast with the behavior of the Young's modulus (E) anomaly across T_M observed by Makkay, Geiger, and Fine. We conclude that the v_p anomaly is intrinsic to the saturated crystal.

INTRODUCTION

Our purpose in this paper is to report measurements of the isotropic elastic properties of hematite as a function of temperature in the region of the Morin[1] transition (T_M), to give an acoustic determination of ($\partial T_M/\partial P$), and to discuss the origins of the observed elastic behavior in terms of models proposed by previous investigators.

EXPERIMENTAL RESULTS

Our polycrystalline specimen of hematite was fabricated using a hot press; the bulk density was determined to be 5.254 g/cm³ or 99.6% of the x-ray density.[2]

The pressure and temperature dependence of the elastic properties near room temperature have been reported previously for this specimen.[2,3] The single crystal of hematite used in this study was kindly lent by Y. Shapira and is part of the same Brazilian crystal employed in his recent[4] studies; this crystal has a low-impurity content and exhibits sharp magnetic phase transitions.

The pulse superposition method[5] was employed to obtain the pulse repetition frequencies (PRF), or reciprocal transit times, which are directly proportional to the elastic-wave velocities. The precision of the PRF measurements by this method is 10 ppm. The measurements of the PRF for the compressional and shear modes were made with X-cut (40 MHz) and Y-cut (30 MHz) quartz transducers, respectively, which were bonded to the specimens with Dow-Corning resin 276-V9.

In Fig. 1 the PRF is plotted as a function of temperature for the compressional and shear modes in the hematite polycrystal. Since the length change[6] over the temperature range from 200° to 300°K is less than 6 ppm, Fig. 1 is essentially a plot of velocity vs temperature. At $P=1$ bar, both the compressional and shear modes exhibit an anomalous behavior between 230° and 270°K.

The Morin transition appears to have a more pronounced effect upon the shear than upon the compressional mode. While T_M is difficult to define on the basis of the velocity data alone, the shear mode has a minimum near 253°K at P=1 bar; in the absence of an applied magnetic field, the Morin transition for the polycrystalline specimen appears to increase by about 4°C under a pressure of 15 000 psi (1.03 kbar). The vertical arrows on the curve correspond (in order of decreasing temperature) to 90%, 50%, and 10% values of the weak ferromagnetic moment on the remanence versus temperature curve for the polycrystalline specimen.[7] Thus the midpoint of the remanence curve and the minimum in the shear mode differ by about 2°C. The shear velocity at T_M was changed by less than 0.02% when the experiment was performed in an applied field of 650 Oe. The coercive force for the same specimen was found to be 270 Oe.

To verify our determination of $(\partial T_M/\partial P)$ and to test whether the observed anomalies in the compressional (v_p) and shear (v_s) modes in the polycrystal have their origin in domain wall-stress interaction in a magnetically saturated crystal, v_p was measured along the trigonal (c) axis of the single crystal of hematite. It will be recalled that above T_M the magnetic spins in hematite are wholly contained in the c plane and the c axis is the hard magnetic axis. As illustrated in Fig. 2, v_p exhibits a sharp increase ($\Delta v_p=0.2\%$) as the crystal was cooled through $T_M=(261.0\pm0.2)$°K at $P=1$ bar. The transition occurs very rapidly (within 0.4°K), exhibits a small amount of hysteresis (<0.2°K) upon heating, and is consistent with Shapira's[4] determination of $T_M=(260.6\pm0.5)$°K. The v_p jump was unaffected by 650 Oe applied in the c plane. The coercive force for this specimen was less than 3 Oe.[7]

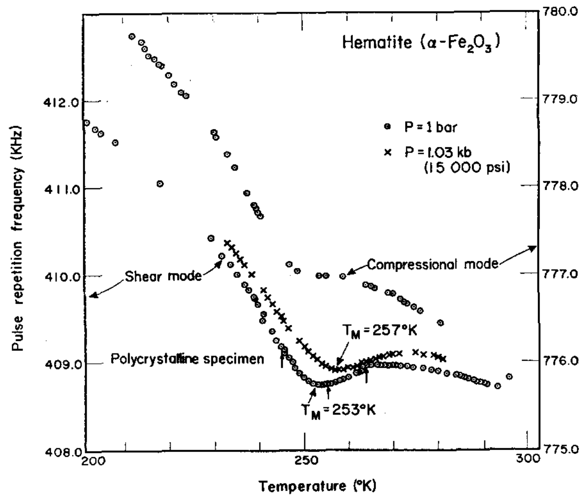

FIG. 1. Pulse repetition frequency versus temperature for compressional and shear modes in polycrystalline hematite from 200° to 300°K at $P=1$ bar. Measurements also given for shear mode at $P=1.03$ kbar (15 000 psi).

The abrupt jump in v_p at T_M in the single crystal makes possible a much more precise determination of $(\partial T_M/\partial P)$. An automatic peak finder designed by P. Mattaboni enables us to lock on the pseudo-resonant PRF and to follow rapid changes, such as those depicted in Fig. 2, and thus to determine the midpoint of the transition at each pressure. From the data in Fig. 2, we see that T_M is linear in P between 1 bar and 1 kbar and that $(\partial T_M/\partial P)=(3.6\pm0.4)$° K/kbar.

DISCUSSION

It is interesting to compare our acoustic determination of $(\partial T_M/\partial P)$ with the results of previous investigators using the techniques of magnetic measurements, neutron diffraction, and nuclear magnetic resonance (NMR). Our measurement of $(\partial T_M/\partial P)=(3.6\pm0.4)$°K/kbar is in agreement with that determined from neutron diffraction [8,9] and NMR studies[10] [$(\partial T_M/\partial P)=(3.7\pm0.3)$°K/kbar] but differs from that measured by Kawai and Ôno[11] [$(\partial T_M/\partial P)=10$°K/kbar].

The sharp increase ($\Delta v_p=0.2\%$) observed as the single crystal was cooled through T_M did not vanish when the experiment was repeated with a magnetic field of 650 Oe (two orders of magnitude greater than the coercive force) applied in the c plane; nor is there any difference in attenuation of the acoustic signal above or below T_M. This is in marked contrast with the behavior of the Young's modulus (E) anomaly along the c axis observed by Makkay *et al.*[12]; their ΔE

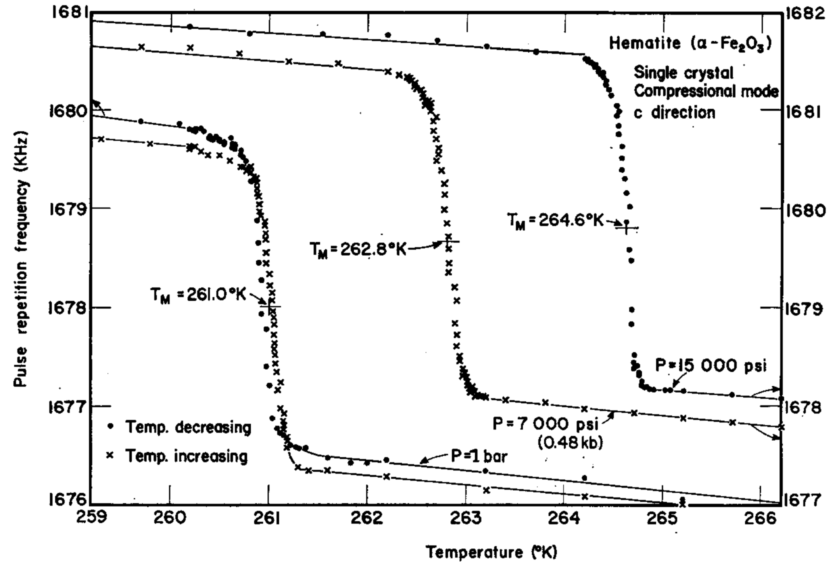

FIG. 2. Pulse repetition frequency versus temperature in the region of the Morin transition (T_M) for compressional waves propagating along the trigonal (c) axis of hematite for 3 different hydrostatic pressures (P).

anomaly was removed by the application of a 1570-Oe field in the c plane. Thus, if the domain wall-stress interaction (ΔE effect) is to be the explanation of our velocity jump, then hematite must have a high H_c (>650 Oe) moment below T_M. Because magnetic measurements do not confirm this, we believe rather that the v_p anomaly is intrinsic to the saturated crystal and is very likely the consequence of spin-wave-phonon interaction. Although the spin-wave-phonon interaction for compressional waves should be zero for particle vibration parallel or perpendicular to the spin axis, the interaction is nonzero and finite for small angles [5° to 22° for YIG [13]] between the spins and particle vibrations. Morrish *et al.*[14] have concluded from neutron diffraction data that below T_M the spins are inclined by about 10° to the c axis (our propagation and particle vibration direction). We suggest, therefore, that the observed v_p anomaly is due to the presence of a linear spin-wave-phonon interaction below T_M.

Note added in proof: Subsequent work[15] on the single-crystal hematite indicates that the sharp increase in v_p at T_M vanishes if the experiment is performed with a magnetic field of 4 kOe applied in the c-plane. Thus all of the effects observed in this paper may be explained by domain wall–stress interaction without appealing to spin wave–phonon interactions. It is important to note that saturating fields which are several orders of magnitude greater than the observed H_c are required to suppress domain wall–stress interactions in hematite at room temperature.

ACKNOWLEDGMENTS

We are grateful to O. L. Anderson for his enthusiastic support of this work, to Y. Shapira for the loan of his single crystal of hematite, to P. Mattaboni for technical assistance, and to the National Science Foundation for financial support.

* Lamont-Doherty Geological Observatory Contribution No. 1415.

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