# Elastic and Magnetoelastic Effects in Magnetite*

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The temperature dependence of the longitudinal and the shear sound-wave velocity along the [100] axis in $Fe_3O_4$ is presented. The longitudinal velocity exhibits an anomaly at 130°K, where there is an easy-axis change from the [111] to the [100]. Both shear and longitudinal velocities show sharp increases of $7\frac{1}{2}\%$ and 0.2%, respectively, at 119°K, where the structure changes from cubic to orthorhombic. In addition, the linear magnetoelastic birefringence effect and the shear-velocity changes as a function of magnetic field were used to determine the magnetoelastic coupling constant $b_{44}$ as a function of temperature from room temperature to 125°K. These results are compared with values of $b_{44}$ obtained from magnetostriction data. Transverse ferroacoustic resonance effects could be observed in the temperature interval 150–300°K.

## I. INTRODUCTION

IN the course of a general investigation of magnetic materials using ultrasonic techniques, we have studied the elastic and magnetoelastic properties of magnetite ($Fe_3O_4$). The static properties of $Fe_3O_4$, such as magnetocrystalline anisotropy, magnetostriction, thermal expansion, etc., have been investigated extensively.[1] Furthermore, its cubic structure in the high-temperature phase (above 119°K) and the rather large values of the magnetoelastic coupling constants make $Fe_3O_4$ an attractive substance to investigate ultrasonically. Its magnetoelastic properties at room temperature have previously been studied by one of us[2] using the so-called linear birefringence effect. This work extends these measurements down to the phase transition at 119°K and also includes high-resolution velocity measurements (1:$10^6$) as a function of temperature and for different geometric configurations of the magnetization and sound polarization with propagation direction along the [100] axis.

In Sec. II we shall present the experimental details and in Sec. III the theory of the magnetoelastic effects. In Sec. IV we will present the experimental data, which include the temperature dependence of the shear and longitudinal sound velocity along the [100] axis and velocity and attenuation measurements as a function of magnetic field. Values of the magnetoelastic coupling constant $b_{44}$ obtained from these data will be compared with those obtained from magnetostriction measurements.[3]

## II. EXPERIMENTAL DETAILS

The birefringence data were obtained using standard pulse-echo techniques. The amplitudes of single echoes were automatically recorded as a function of magnetic field using a peak detector, and oscillatory patterns such as the one shown in Fig. 1 could be obtained.

The velocity measurements were made using a phase comparison technique similar to one used by Eastman.[4] A block diagram of the system is shown in Fig. 2. A Hewlett-Packard model 608F signal generator coupled with a model 8708A synchronizer provides a cw-rf signal stable to 1:$10^7$ in frequency. This stable source is then split into two channels, one of which is gated by a diode switch to form pulses which are then amplified, injected into the sample, and the echo train received. At this point both the cw reference and the echoes are beat down to 30 MHz in separate mixers by the same local oscillator. This mixing step leaves the phase relationship of the two signals unchanged. Both channels are then amplified, with the cw reference passing through a 75-nsec delay line before amplification. They are then mixed in a Hewlett-Packard 1015A balanced mixer which acts as a phase detector. Its output is then amplified and the interference echo pattern viewed on an oscilloscope.

Velocity changes are measured by bringing a single echo's phase to null by adjustment of either the source frequency or the delay line. After a velocity or transit-

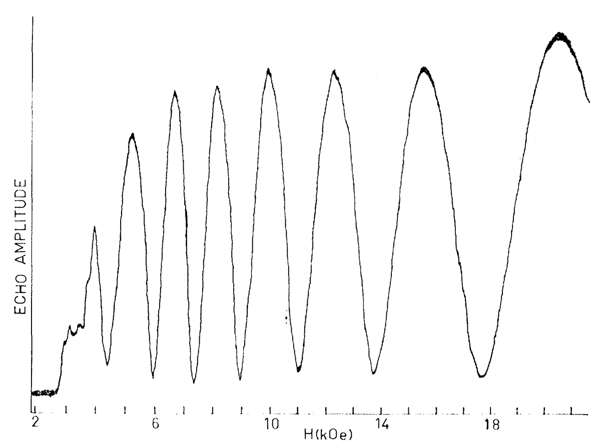


FIG. 1. Typical $X$-$Y$ recorder plot of the amplitude of a single echo versus magnetic field: $\nu=150$ MHz (echo 10), $T=160$°K. **R**, [001]; **H**, [011].

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[1] L. R. Bickford, Jr., Rev. Mod. Phys. **25**, 75 (1953). More recent literature on $Fe_3O_4$ is included in E. Callen, Phys. Rev. **150**, 367 (1966).
[2] B. Lüthi, Appl. Phys. Letters **8**, 107 (1966).
[3] L. R. Bickford, Jr., Phys. Rev. **99**, 1210 (1955).
[4] D. E. Eastman, Ph.D. thesis, MIT, 1965 (unpublished); Phys. Rev. **148**, 530 (1966).

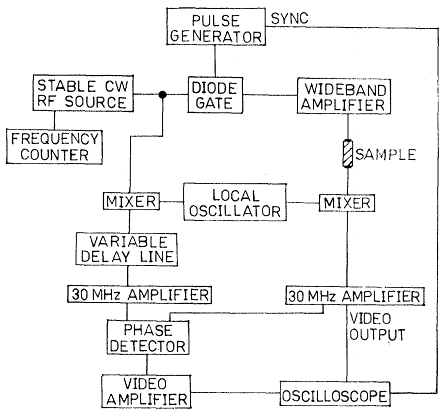


FIG. 2. Block diagram of experimental setup.

time change the selected echo's phase is again brought to null by adjusting the frequency. The change in acoustic transit time $T_L$ is given by $\Delta T_L/T_L=-\Delta f/f=-\Delta v/v+\Delta l/l$ if $T_{\text{electron}}\ll T_{\text{acous}}$.

Changes of 1:$10^6$ were observable with the acoustic delay available in our present case. With longer delays, 1:$10^7$ resolution is possible. This system's main advantage over others of similar resolution is its capability of measuring frequency-dependent velocity effects. This feature was not used in the present investigation, but we have found it invaluable in several others.[5]

The accuracy of the system is affected by several factors. Foreign modes in the echo pattern and amplitude variations will cause spurious apparent velocity changes. Care must also be taken to remain within the bandwidths of all the components of the system.

The crystal used was the same one of Ref. 2. It is of cylindrical form with its axis oriented along a $\langle 100\rangle$ direction.

## III. THEORY

The linear theory of magnetoelastic interaction has been given first by Kittel[6] and by Akhiezer.[7] It has been worked out in detail by Schlömann.[8] Linear magnetoelastic interaction occurs in the region where the phase velocities of the spin waves and phonons are the same. We are interested in the case where propagation direction $\mathbf{k}$ and saturation magnetization $\mathbf{M}_0$ are mutually perpendicular. With $\mathbf{k}$ parallel to [100] and $\mathbf{M}$ in the (100) plane, the following relations hold[8,9]:

$\mathbf{R}\|\mathbf{M}$, coupled modes:

$$(\omega^2-v^2k^2)(\omega^2-\omega_m^2)=\sigma\gamma H_i v^2k^2\,,$$

$\mathbf{R}\perp\mathbf{M}$, uncoupled modes:

$$(\omega^2-v^2k^2)(\omega^2-\omega_m^2)=0\,. \qquad (1)$$

Here $\sigma=\gamma b_{44}^2/\rho M_0 v^2$ is a measure of the strength of the interaction. The spin-wave frequency $\omega_m$ has the following form for $M_0$ parallel to a cube edge[9]:

$$\begin{aligned}\omega_m^2=&\gamma^2(H-NM_0+2K_1/M+\alpha k^2)\\&\times(H-NM_0+4\pi M_0+2K_1/M_0+\alpha k^2)\,,\quad (2)\\H_i=&H-NM_0+2K_1/M_0\,.\end{aligned}$$

For our frequency range, the exchange term $\alpha k^2$ is dominated by the other terms in the expression and may be neglected.

From Eq. (1) we can easily calculate the group and phase velocities $v_g$ and $v_p$ in the coupled-mode geometry. For $\omega_m\gg\omega$ and $\omega_m\gg\sigma$, we find $v_g=v_p$ and

$$-\Delta v/v=\gamma H_i\sigma/2\omega_m^2\,. \qquad (3)$$

Similar expressions have been derived by Eastman.[4]

By measuring the change in sound velocity versus magnetic field for these geometries one can determine the coupled-mode dispersion spectrum, e.g., one can determine relevant parameters such as magnetoelastic coupling constant $b_{44}$ and anisotropy fields, etc.

Shear-wave propagation along a [100] direction is elastically isotropic in a nonmagnetic material, e.g., independent of the polarization direction $\mathbf{R}$ in the [100] plane. As seen from Eqs. (1) and (3), the spin-wave–phonon coupling in a magnetic material introduces an anisotropy. The sound velocity depends on the orientation of $\mathbf{R}$ with respect to $\mathbf{M}_0$. The two normal modes of propagation are $\mathbf{R}\|\mathbf{M}_0$ (coupled) and $\mathbf{R}\perp\mathbf{M}_0$ (uncoupled). Thus the medium becomes doubly refracting. We call this effect linear magnetoelastic birefringence.[2] For $\mathbf{R}$ at 45° to $\mathbf{M}_0$ the two components of $\mathbf{R}$ parallel and perpendicular to $\mathbf{M}_0$ experience different phase velocities. If the phase difference in the two components is $\frac{1}{2}(2n+1)\pi$, the originally linearly polarized sound wave becomes circularly polarized, and for $n\pi$ it is linearly polarized. With a linearly polarized quartz transducer as a receiver, this developing phase difference can be measured by measuring the apparent attenuation either as a function of distance or magnetic field (Fig. 1). The phase difference per unit length is (for $\omega_m\gg\omega$)[2]

$$\phi/l=(\sigma\omega/2v)\gamma H_i/\omega_m^2\,. \qquad (4)$$

For $M_0$ along [011], $\mathbf{R}$ along [001], and $\mathbf{k}$ parallel to [100],[2]

$$\begin{aligned}\phi/l=&(\omega\sigma/2v\gamma)(H-NM_0+4\pi M_0\\&+K_1/M+K_2/2M)^{-1}\,. \qquad (5)\end{aligned}$$

From the birefringence effect one gets the same kind of information as from the velocity changes in the coupled-mode geometry. Comparing Eqs. (3) and (4),

[5] B. Lüthi and R. J. Pollina, Phys. Rev. Letters **22**, 717 (1969); and to be published.
[6] C. Kittel, Phys. Rev. **110**, 836 (1958).
[7] A. I. Akhiezer, V. G. Bariakhtar, and S. V. Peletminski, Zh. Eksperim. i Teor. Fiz. **35**, 157 (1958) [English transl.: Soviet Phys.—JETP **8**, 157 (1959)].
[8] E. Schlömann, J. Appl. Phys. **31**, 1647 (1960).
[9] B. Lüthi and F. Oertle, Physik Kondensierten Materie **2**, 99 (1964).

one finds the following simple relation:

$$-(v/\omega)\phi/l=\Delta v/v. \tag{6}$$

This expression is valid even for two different directions of $\mathbf{M}$ as long as $2K_1/M_0$ and $K_2/2M_0$ are small, in which case $\omega_m$ is practically independent of the direction of $\mathbf{M}$, as long as $\mathbf{M}$ is perpendicular to $\mathbf{k}$.

The dispersion effects treated in Eqs. (1)–(4) are accompanied by an attenuation effect observable in the coupled-mode geometry. We call it transverse ferroacoustic resonance.[9] It can be phenomenologically described by introducing a spin-wave relaxation time, e.g., by substituting $\omega_m+i/\tau$ for $\omega_m$, although the relaxation mechanism may be of a different nature. From Eqs. (1) and (2) it follows that the resonance field for $M_0$ along an [001] direction is given by

$$H_r \simeq NM_0-2K_1/M_0+\omega/4\pi\gamma M_0. \tag{7}$$

For our frequencies $\omega/4\pi\gamma M_0$ is negligible. Then, according to Néel's phase model, $H_r$ is just the field necessary to saturate the sample. We therefore expect to observe this ferroacoustic resonance only under very favorable conditions.

## IV. RESULTS AND DISCUSSION

### A. Elastic Results

Figure 3 shows the temperature dependence of the longitudinal sound velocity propagated along the [100] direction. The lower curve was obtained in zero applied field. The velocity exhibits a normal increase with decreasing temperature until approximately 200°K, where it levels off and then decreases slightly. Just below 140°K there is a sudden decrease, with the minimum in the vicinity of 130°K, where the easy direction of magnetization changes from the [111] to the [100].[1] Accompanying this change is a large increase in the attenuation which resulted in a loss of the echo pattern just above 131°K. This prevented an exact determination of the extent of the velocity change at this point. When measurement was resumed at 127°K, a new reference value for the velocity had to be chosen (see below), since we could not measure the absolute velocity to the required accuracy. Below the easy-axis change we find a sudden jump of about 0.2% (after correction for thermal expansion) at 119°K, where the structure changes from cubic to orthorhombic.[1] We found that application of a magnetic field along the [001] direction caused an increase in velocity until the saturation field was reached for temperatures above 119°K, after which the velocity remained constant. The upper curve represents the velocity in a saturating field. The displacement of the two curves is the change resulting from going to the magnetized state from the demagnetized one. It can be seen that

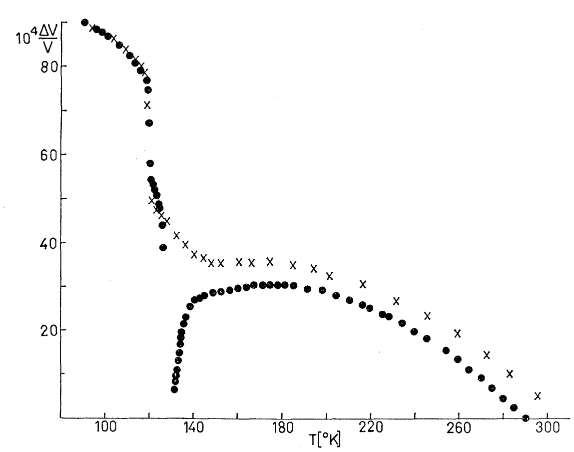


FIG. 3. Temperature dependence of the longitudinal sound velocity along [100] axis: ●, $H=0$; ×, $H=10$ kOe; $\nu=50$ MHz.

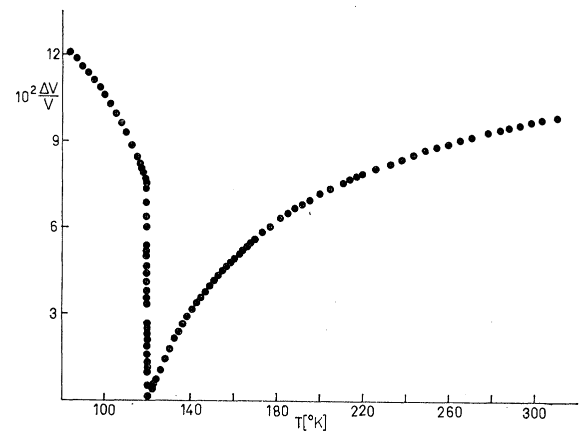


FIG. 4. Temperature dependence of the shear sound velocity along [100] axis: **R** along [001]; $H=15$ kOe along [010]; $\nu=50$ MHz.

the large change at 130°K has been effectively removed by the field, since the sample remains saturated. The jump at the phase transition remains unchanged in magnitude. Below the transition it was found that the velocity was practically independent of field and the two curves coincide in this region. This is expected to be the case, since cooling the sample through the transition with a saturating field applied along an axis fixes the easy direction along that axis, eliminating domain-wall scattering effects, etc.

Figure 4 shows the temperature dependence of the velocity of a shear sound wave propagating along the [100] direction with polarization along the [001]. The zero-field attenuation was so large in this case that a saturating field was needed to observe an echo pattern. The field was applied in the [010] direction, since from Eq. (1) there is no spin-wave–phonon coupling in this geometry. The velocity was independent of field above saturation as expected. It can be seen that the velocity decreases down to 119°K, where we find an extremely large jump of almost $7\frac{1}{2}\%$, in contrast to the 0.2% in the longitudinal case.

Since the transition at 119°K is noticeably of first order, no critical velocity and attenuation effects can be observed at this transition.[10] From the velocity changes in Figs. 3 and 4, one gets the elastic constants $C_{11}$ and $C_{44}$ by means of $v_L=(C_{11}/\rho)^{1/2}$ and $v_S=(C_{44}/\rho)^{1/2}$, where $\rho=5.185$ and, at 300°K, $v_L=6.75\times10^5$ cm/sec and $v_S=3.69\times10^5$ cm/sec.

### B. Magnetoelastic Effects

Figure 5 shows the phase curves $\phi/l$ as a function of $H$ [Eq. (5)], obtained at temperatures from 220 to 125°K by plotting the field values of the maxima and minima of individual echoes (Fig. 1) as a function of phase change per unit length. The data obtained at 70 MHz have been scaled to those at 150 MHz, using the frequency dependence predicted by Eq. (5). The solid lines represent a theoretical fit using the same equation. The predicted field and frequency dependences are well obeyed over a wide field range in the saturated region.

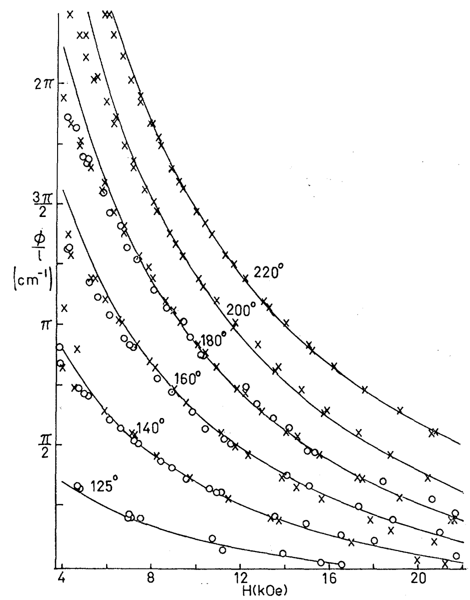


FIG. 5. Linear magnetoelastic birefringence: ×, 150 MHz; ○, 70 MHz; solid curves, theoretical fit.

Figure 6 shows the field dependence of the velocity of the coupled mode from 260 to 130°K. The velocity of the uncoupled mode was found to remain constant with field above saturation. The solid lines again represent a theoretical fit of the data [Eq. (3)] using the same values of effective field and spin-wave frequency as were used for the phase curves. Again the fit is quite good.

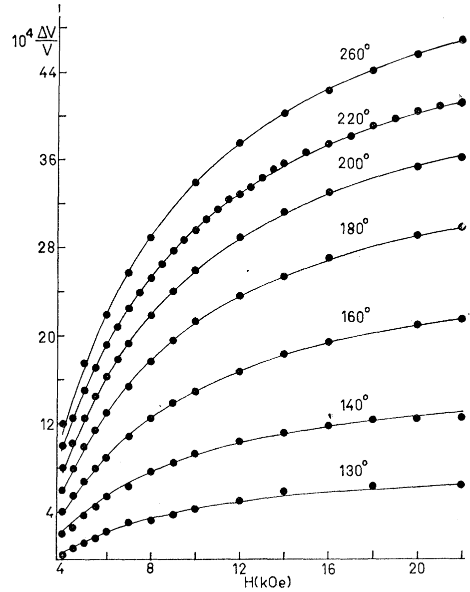


FIG. 6. Magnetic field dependence of the shear sound velocity: **k**, [100]; **R**, $H$ [001]; solid curves, theoretical fit.

In Fig. 7, we have plotted the $b_{44}$ values which gave the best fit to the data in Figs. 5 and 6. Calculated values for the demagnetizing field and published values of the magnetization and anisotropy energies were used in Eqs. (3) and (5) when $b_{44}$ was calculated. The solid line is the coupling constant obtained from magnetostriction data.[3] The agreement between velocity and birefringence measurements is quite good and they have approximately the same temperature dependence as the magnetostriction data. The divergence at low temperatures between our data and the magnetostriction results can be attributed to different samples and

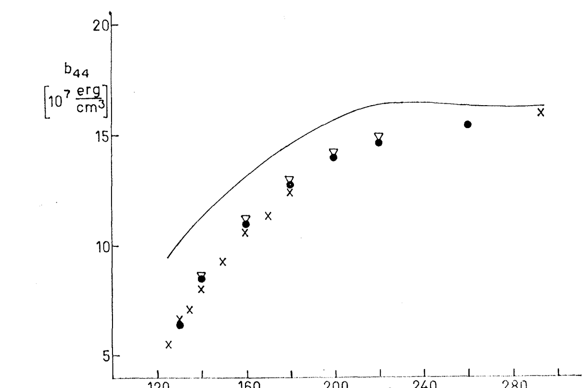


FIG. 7. Temperature dependence of $b_{44}$: ×, 70-MHz birefringence data; ▽, 150-MHz birefringence data; •, velocity data; solid curve, magnetostriction data (Ref. 3).

[10] For a discussion of these critical effects, see, e.g., B. Lüthi, P. Papon, and R. J. Pollina, J. Appl. Phys. **40**, 1029 (1969).

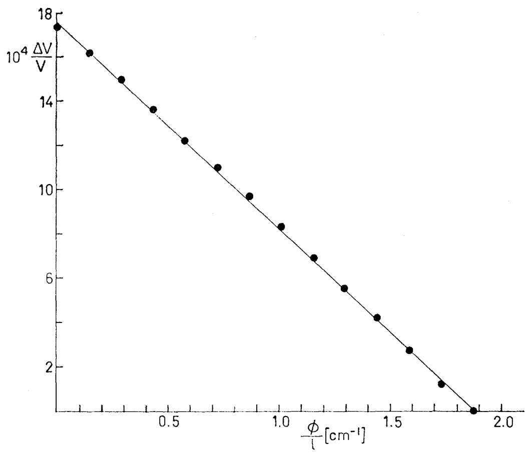


FIG. 8. Typical plot of $\Delta v/v$ versus $\phi/l$: $T=160°$K, $\nu=70$ MHz.

to the fact that the magnetostriction data are claimed accurate only to $\pm 20\%$ at these temperatures.

Figure 8 shows a typical plot of $\Delta v/v$ versus $\phi/l$. It can be seen that there is a linear relationship as predicted [Eq. (6)], and the sound velocity obtained from the slope of the line agrees to within 10% with the measured velocity.

The transverse ferroacoustic resonance effect[9] was found for temperatures down to 150°K in an unsaturated field region, which made the exact field value difficult to determine (see Fig. 9). The field values of the resonance agreed approximately with calculated values [Eq. (7)], taking the same values for the anisotropy energy and magnetization as in the birefringence and velocity calculations. Below 150°K the resonance was very damped and could not be resolved with any accuracy. The linewidth of this resonance probably arises from the inhomogeneous demagnetizing field[2] and not from intrinsic spin-wave relaxation mechanisms. For the uncoupled geometry we did not observe any such resonance, in agreement with Eq. (1).

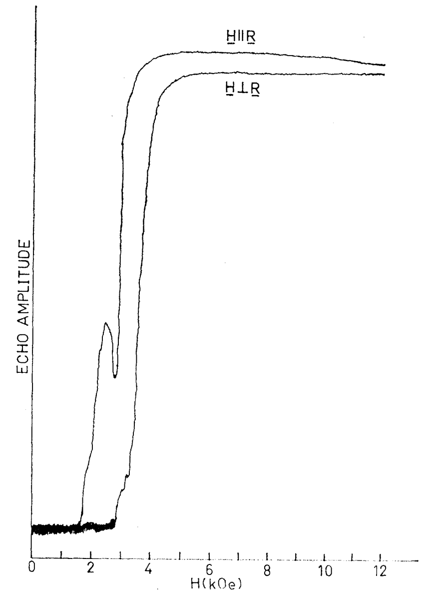


FIG. 9. Typical $X$-$Y$ recorder plot of the transverse ferroacoustic resonance for $H\|R$. No resonance effect is seen for $H\perp R$. $T=180°$K; $\nu=70$ MHz.

## V. CONCLUSION

This investigation shows that in cubic materials the linear phonon–spin-wave effects are very well understood. The different magnetoelastic effects discussed and demonstrated have been quantitatively understood and are interrelated. As we shall see in subsequent publications, other materials such as rare-earth metals are much more complex and more difficult to interpret quantitatively.[11]

In addition, we have shown from our velocity measurements that elastic constants are greatly affected by changes in order (easy-axis changes at 130°K and first-order phase transition at 119°K).

[11] T. J. Moran and B. Lüthi, J. Phys. Chem. Solids (to be published).