

Parallel Pumping of Magnetoelastic Waves in Lithium Ferrite

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The parallel pumping technique is used to measure the magnetoelastic properties of single-crystal lithium ferrite at room temperature. This technique is extended to cover measurements of the magnetoelastic coupling constant B_1 . Measurements of the exchange constant, spin-wave linewidth, and elastic Q at 17.3 Gc/sec are also presented. Molecular-field theory predicts an exchange constant for lithium ferrite which is half the experimental value. The three-magnon relaxation process accounts for only part of the k dependence of the spin-wave losses. The elastic Q for lithium ferrite at this frequency is approximately half that for yttrium iron garnet.

INTRODUCTION

IT has been shown by Schlömann^{1,2} and Morgenthaler³ that spin waves can be parametrically excited by an rf magnetic field applied parallel to the saturating magnetic field in ferromagnetic insulators.¹⁻³ Turner,⁴ following Schlömann's prediction,¹ demonstrated, using yttrium iron garnet (YIG), that coupled spin and elastic waves (magnetoelastic waves) also can be "parallel pumped." The presence of these waves in Turner's experiment was evidenced by an increase in the parallel pumping instability threshold when the dc magnetic field was in the vicinity of the value corresponding to a "crossover" of the $\pi/2$ spin-wave and elastic-wave spectra. The dc magnetic field for the increase in the instability threshold due to elastic-wave coupling can be used to measure the exchange field acting on the $\pi/2$ spin waves and this field was measured for YIG.^{4,5} Morgenthaler⁶ showed that the height and width of the threshold rf field variation versus dc magnetic field curve in the interaction region (the "notch") can be used to determine one of the magnetoelastic coupling constants (B_2) and the elastic Q of the magnetoelastic wave when the magnetic field is along [100]. This technique has been used to measure B_2 and the elastic Q as a function of temperature for YIG.⁷

This paper presents similar experimental results for single-crystal lithium ferrite at 300°K with the magnetic field along [111]. Previous measurements^{8,9} had established that the magnetoelastic coupling constant of lithium ferrite was an order of magnitude larger than for YIG, with B_1 larger by an order of magnitude than B_2 , and that the linewidth was comparable (as

low as 1 Oe⁹). A detailed investigation of the magnetoelastic properties was thus clearly of interest. In order to measure B_1 it is necessary to use a different orientation of the dc magnetic field than [100] since in this case only B_2 is involved in the interaction of shear elastic and spin waves.⁷ As discussed in Appendix A, if the sample is oriented with the magnetic field along the [111] (easy) axis, the measurement yields a combination coupling constant of $(2B_1+B_2)/3$. A comparison is made between the spin-wave linewidths and the three-magnon relaxation calculation of Sparks, Loudon, and Kittel.¹⁰

The lithium ferrite samples used in the experiments were polished spheres approximately 1 mm in diameter. They were flux grown by a process described in Ref. 11. All measurements were done with the sample in the ordered state, which was found to result in the lowest parallel pumping threshold.¹² The experiments were carried out at a pump frequency (ω_p) of 34.67 Gc/sec, using a full wavelength critically coupled reflection cavity. This high frequency was used for two reasons. First, it ensured magnetic saturation of the sample over the magnetic fields of interest. Second, the longitudinal and shear-wave notches are widely separated at this frequency. The samples were mounted in the cavity loosely in a Teflon holder so as to be free to rotate. The ferromagnetic resonance linewidth of the particular sample used in these experiments was 5 Oe at 6.2 Gc/sec. A large part of the resonance losses was probably caused by the imperfect surface polish which should not contribute to the parallel pumping instability threshold. The presence of the instability was detected by observing the change in the reflected power as the dc magnetic field was varied. Above the instability threshold low-frequency relaxation oscillations were observed which were similar to those previously observed for YIG.¹³ The rf magnetic susceptibility of the parallel-pumped instability was always smaller compared to that observed in similar experiments with YIG.

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¹ E. Schlömann, Technical Report R-48, Research Division, Raytheon Company, 1959 (unpublished).

² E. Schlömann, J. J. Green, and U. Milano, J. Appl. Phys. **31**, 386S (1960).

³ F. R. Morgenthaler, Suppl. J. Appl. Phys. **31**, 95S (1960).

⁴ E. H. Turner, Phys. Rev. Letters **5**, 100 (1960).

⁵ R. C. LeCraw and L. R. Walker, Suppl. J. Appl. Phys. **32**, 167S (1961).

⁶ F. R. Morgenthaler, J. Appl. Phys. **34**, 1289 (1963).

⁷ F. A. Olson, J. Appl. Phys. **34**, 1281 (1963).

⁸ S. Iida, Bell Telephone Laboratories (unpublished work).

⁹ R. L. Comstock and J. P. Remeika, J. Appl. Phys. **35**, 1018 (1964).

¹⁰ M. Sparks, R. Loudon, C. Kittel, Phys. Rev. **122**, 791 (1961).

¹¹ J. P. Remeika and R. L. Comstock (to be published).

¹² R. T. Denton and E. G. Spencer, Suppl. J. Appl. Phys. **33**, 1300 (1962).

¹³ T. S. Hartwick, E. R. Peressini, and M. T. Weiss, Suppl. J. Appl. Phys. **32**, 223S (1961).

TABLE I. Magnetic and elastic properties of lithium ferrite and yttrium iron garnet at 300°K.

	Lithium ferrite	YIG
$4\pi M$ (G)	3640	1770
$v_l \times 10^{-5}$ (cm/sec)	7.98 ^a	7.21([100]), 7.15([110])
$c_{11} \times 10^{-11}$ (dyn cm ⁻²)	30.2	26.9 ^b
$v_s \times 10^{-5}$ (cm/sec)	3.78 ^a	3.84 ^b
$c_{44} \times 10^{-11}$ (dyn cm ⁻²)	6.77	7.64 ^b
$D \times 10^9$ (Oe cm ²)	8.81	5.17 ^c
$B_1 \times 10^{-6}$ (erg cm ⁻³)	68.5	3.48 ^d
$B_2 \times 10^{-6}$ (erg cm ⁻³)	$\ll B_1$	6.96 ^d
$Q_c(17.3 \text{ Gc/sec})$	5300	12 000 ^e
$\Delta H_{k \rightarrow 0}$ (Oe)	3.52 at 17.3 Gc/sec	0.205 at 5.7 Gc/sec ^f
$\partial \Delta H_k / \partial k \times 10^8$ (Oe cm)	0.92 at 17.3 Gc/sec	1.19 at 5.7 Gc/sec ^f
$H_{\text{anis.}}([111])$ (Oe)	450(300°K), ^h 664(4.2°K) ^h	53(300°K), ^g 165(4.2°K) ^g
g_{eff}	1.958(300°K) ^h 1.962(4.2°K) ^h	2.005(300°K) ^g 2.001(1.5°K) ^g

^a Longitudinal and shear-wave velocities (v_l and v_s) measured on an unmagnetized disk by H. J. McSkimin and v_s measured by R. C. LeCraw on a saturated sphere with the same result.

^b All elastic constants from A. E. Clark and R. E. Strakna, J. Appl. Phys. 32, 1172 (1961).

^c Reference 5.

^d A. B. Smith and R. V. Jones, J. Appl. Phys. 34, 1283 (1963).

^e Linear extrapolation from Ref. 7.

^f R. C. LeCraw and E. G. Spencer, J. Phys. Soc. Japan 17, Suppl. B-1, 401 (1962).

^g G. P. Rodrigue, H. Meyer, and R. V. Jones, Suppl. J. Appl. Phys. 31, 376S (1960).

^h A. D. Schnitzler, V. J. Folen, and G. T. Rado, Suppl. J. Appl. Phys. 33, 1293 (1962), measured $H_{\text{anis}}([111])$ and g_{eff} on less stoichiometric lithium ferrite. They found $H_{\text{anis}}([111])$ equal to 382 at 300°K and 865 at 4.2°K. The value of g_{eff} was greater than 2 from 4.2°K to room temperature.

EXPERIMENTAL RESULTS

In Fig. 1 is shown the experimental curve of the threshold for the instability as a function of $(|H_c - H|)^{1/2}$. Here, H_c is the external dc magnetic field corresponding to the minimum observed threshold for excitation of spin waves with $k \rightarrow 0$ (k is the wave number). The ordinate is the threshold normalized to the minimum threshold, $h_{\text{crit, min}}$. The "notches" in this curve correspond to the excitation of magnetoelastic waves with longitudinal (H_1) and shear (H_2) elastic components, both of which propagate at right angles to the dc magnetic field. The longitudinal notch was not observed in the experiments described in Ref. 7 since longitudinal waves are not coupled to spin waves when the dc magnetic field is along [100] (see Appendix A). The wave number of the $\pi/2$ spin waves for $H < H_c$ is proportional to the abscissa, i.e.,

$$k = [(H_c - H)/D]^{1/2}, \quad (1)$$

where D is the exchange field constant.¹⁴ The parameter D is defined by the dispersion relations for $\pi/2$ spin waves in a sphere as

$$(\omega_p/2\gamma)^2 = (H_{\text{eff}} - (4\pi M/3) + Dk^2) \times (H_{\text{eff}} + (8\pi M/3) + Dk^2), \quad (2)$$

where $4\pi M$ is the saturation magnetization (3640 G at 300°K⁹) and γ is the gyromagnetic ratio. The value of γ measured on our sample of lithium ferrite at 300°K was $2\pi \cdot 2.740$ (Mc/sec Oe)¹⁵ (see Table I). The effective field (H_{eff}) includes the anisotropy field, which is 449 Oe at 300°K as measured along [111].¹⁵ The assignment of the two notches can be made on the basis of

¹⁴ The connection between the parameter D used here and that used in Ref. 5, referred to here as D' , is $D = D'/\gamma\hbar$, while the relationship to the field used in Ref. 6 is $D = \lambda\omega_m/\gamma$.

¹⁵ R. L. Comstock, W. G. Nilsen, and J. P. Remeika (to be published).

the known wave velocities. From Eq. (1) and the dispersion relation for elastic waves, the position of the notches should be given by

$$[(H_c - H_2)/(H_c - H_1)]^{1/2} = v_l/v_s, \quad (3)$$

where v_l and v_s are the longitudinal and shear-wave velocities, respectively. From the measured velocities (see Table I) and Fig. 1 we find Eq. (3) to be satisfied within 10%. The shift in the shear-wave interaction predicted by Morgenthaler⁶ should be negligibly small (less than 1 Oe) in our experiment.

The straight line parts of the curve in Fig. 1 correspond to pure spin-wave excitation. The normalized height of the shear-wave notch is 0.064, while the half-width (see inset) is 38 Oe^{1/2}.

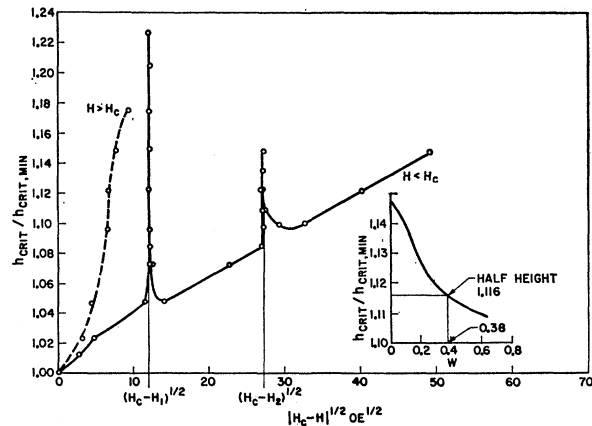


Fig. 1. Parallel pumping threshold at 34.7 Gc/sec normalized to the minimum threshold ($h_{\text{crit, min}} = 12.9$ Oe) versus $(|H_c - H|)^{1/2}$. The dc magnetic field for the minimum threshold was $H_c = 5362$ G. The notches arise from the interaction of spin waves and longitudinal (H_1) and shear (H_2) elastic waves. The inset shows the width of the shear-wave notch.

DISCUSSION

Exchange Field

The exchange field acting on the $\pi/2$ spin wave can be evaluated from the data shown in Fig. 1 by using the position of either the shear-wave or longitudinal-wave notch. Since the shear-wave velocity was known above magnetic saturation it will be used. From Eq. (1) and the shear-wave velocity given in Table I we find $D=8.8\times 10^{-9}$ Oe cm², which may be compared with 5.17×10^{-9} Oe cm² for YIG as determined at 300°K by LeCraw and Walker⁵ using the same technique.

Rado and Folen¹⁶ have determined the molecular field coefficients for lithium ferrite by fitting parameters derived from Néel's theory to measured curves of saturation magnetization versus temperature. These coefficients can be used in Kaplan's¹⁷ dispersion relation for long-wavelength spin waves in the spinel lattice to determine the effective exchange field parameter D . The molecular field coefficients have tetrahedral-tetrahedral, tetrahedral-octahedral, and octahedral-octahedral components given by^{16,18}

$$\frac{J_{TT}}{k_B} = -19.5^\circ\text{K}, \quad \frac{J_{TO}}{k_B} = -24.0^\circ\text{K}, \quad \frac{J_{OO}}{k_B} = -10.6^\circ\text{K}.$$

Kaplan's result for D is given by¹⁷

$$D = \frac{|(11/2)J_{TO}A_S B_S + J_{TT}A_S^2 + 2J_{OO}B_S^2|}{\gamma\hbar|A_S + 2B_S|} a^2, \quad (4)$$

where A_S and B_S are the average values of the spin for the tetrahedral and octahedral sites, respectively, and $2a$ is the lattice constant. For lithium ferrite $A_S = -5/2$, $2B_S = 7.5/2$ and $2a = 9.33$ Å,¹⁹ resulting in $D = 4.4\times 10^{-9}$ Oe cm², which is approximately half the experimental value given above. For YIG, molecular field theory also predicts an exchange field roughly half the observed value.^{5,20} The origin of this disagreement is probably associated with the molecular field parameters rather than with Kaplan's formula. The limitations of these parameters have been discussed by Smart¹⁸ and specifically for lithium ferrite by Prince.²¹

Magnetoelastic Coupling Constant

From Refs. 6 and 7 and Appendix A, and neglecting B_2 , the equation to be used in evaluating B_1 is given by

$$W' = \frac{2B_1}{3} \left[\frac{Dk_s^2}{2c_{44}} \frac{\omega_1^0 \gamma}{\omega_m(2\omega_1^0 + \omega_m)\delta H_2} \right]^{1/2}, \quad (5)$$

where W' is the width at half-height of the shear-wave notch (see the inset in Fig. 1), ω_1^0/γ is the internal field at the crossover, $\omega_m = \gamma 4\pi M$ and $\delta H_2 = H_c - H_2 = Dk_s^2$. With Eq. (5) our data give $B_1 = 68.5\times 10^6$ erg cm⁻³, compared with 65×10^6 erg cm⁻³ measured by the magnetoacoustic resonance comparison technique⁹ and 67×10^6 erg cm⁻³ measured with a strain gauge.⁸

Spin-Wave Linewidth

The value of the spin-wave linewidth corresponding to the minimum threshold ($H = H_c$) is denoted by $\Delta H_{k \rightarrow 0}$ and is given by²

$$\Delta H_{k \rightarrow 0} = (\omega_m/\omega_p) h_{\text{crit}, \text{min}}. \quad (6)$$

The minimum threshold was measured by comparing the threshold to that for a YIG sample, for which $\Delta H_{k \rightarrow 0}$ was known. From this procedure the minimum threshold for lithium ferrite was found to be 12.9 Oe, resulting in a value of $\Delta H_{k \rightarrow 0} = 3.52$ Oe. This value of $\Delta H_{k \rightarrow 0}$ is approximately half the previously reported one (extrapolated to our frequency).¹² This result is evidence in addition to the x-ray measurements of Remeika and Comstock¹¹ that our sample is more ordered than the one measured in Ref. 12.

For $H < H_c$ the excited spin waves have $k \neq 0$ and in this region (6) can be generalized to

$$\Delta H_k = \Delta H_{k \rightarrow 0} + (\partial \Delta H_k / \partial k) k. \quad (7)$$

The straight line segments of Fig. 1 then yield $\partial \Delta H_k / \partial k = 0.9\times 10^{-6}$ Oe cm. The k -dependent part of the spin-wave relaxation resulting from three-magnon processes has been calculated by Sparks *et al.*¹⁰ They find

$$\partial \Delta H_k / \partial k = (\mu^3 k_B T / \hbar^3) (4\pi M / \gamma^2 \omega_0 D),$$

where the first bracket at 300°K is numerically equal to 28.5×10^6 . For lithium ferrite at 34.67 Gc/sec ($\omega_0 \approx \frac{1}{2}\omega_p - \frac{1}{6}\omega_m$ for $\pi/2$ spin waves) and our measured value of D , the theoretical result is

$$\partial \Delta H_k / \partial k = 0.45\times 10^{-6} \text{ Oe cm}.$$

The spin-wave linewidth has a possible error due to the difficulty in detecting the minimum value of the threshold since the susceptibility was small. This threshold uncertainty cannot entirely reconcile the difference between the two values and the three-magnon process does not appear to account for all of the k dependence of the losses.

Elastic Q

The height of the shear-wave notch is directly proportional to the inverse of the elastic Q ⁶

$$(\delta h_{\text{crit}})_{\text{max}} = 2k_s^2 D (2(\omega_1^0/\omega_m) + 1) Q_e^{-1}. \quad (8)$$

¹⁶ G. T. Rado and V. J. Folen, *J. Appl. Phys.* **31**, 62 (1960).

¹⁷ H. Kaplan, *Phys. Rev.* **86**, 121 (1952).

¹⁸ S. Smart, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. III.

¹⁹ J. Smit and H. P. J. Wijn, *Ferrites* (John Wiley & Sons, Inc., New York, 1959), Chap. VIII.

²⁰ R. L. Douglass, *Phys. Rev.* **120**, 1612 (1960).

²¹ E. Prince, *J. Appl. Phys.* **35**, 993 (1964).

The measured value of $(\delta h_{\text{crit}})_{\text{max}}$ (see Fig. 1), results in a value $Q_e = 5300$. Measurements of Q_e for YIG using the same technique at $\frac{1}{2}\omega_p = 11.6$ Gc/sec resulted in $Q_e \approx 18\,000$.⁷ If, as is frequently observed, Q_e^{-1} varies linearly with frequency, a Q_e at 17.34 Gc/sec of 12 000 is predicted for YIG. Lithium ferrite, therefore, has a Q approximately half that for YIG.

As discussed in Ref. 22, for a given value of B_1 and/or B_2 there exists a value of Q_e above which no magnetoelastic notch in the threshold curve should be observed. Instead, for materials satisfying this condition, the spin-wave instability should merge smoothly into the "elastic-wave" instability. For the above value of B_1 the corresponding Q_e at our frequency is 55 000. The observation of a shear-wave notch with $Q_e = 5300$ is consistent with this result.

CONCLUSIONS

The parallel-pumping technique for measuring magnetic and elastic properties of ferrimagnets has been extended to include measurements of the magnetoelastic coupling constant, B_1 , where previously only B_2 could be measured. In materials with comparable values of B_1 and B_2 two measurements will be necessary to separate the two quantities: one with the dc magnetic field along [111], which determines $(2B_1 + B_2)/3$ and one along [100], which determines B_2 . Measurements of the magnetoelastic parameters for lithium ferrite were made with the [111] axis orientation and are summarized in Table I, where the corresponding values for YIG are included for comparison. The exchange field for lithium ferrite was observed to be approximately twice the value obtained using molecular field theory. The magnetoelastic coupling constant agrees closely with values determined by other methods and is an order of magnitude larger than for YIG. The elastic wave Q at 17 Gc/sec is approximately half that for YIG. The k -independent part of the spin-wave losses are an order of magnitude larger than for YIG while the k -dependent parts are comparable. The three-magnon process calculation of Sparks *et al.*,¹⁰ does not appear to account for all of the measured k -dependent part of the relaxation.

ACKNOWLEDGMENTS

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²² R. L. Comstock, J. Appl. Phys. 35, 2427 (1964).

APPENDIX A: MAGNETOELASTIC ENERGY

For the magnetic field along [100], the magnetoelastic free energy is²³

$$U_{\text{me}}^{[100]} = B_1(\alpha_x^2 e_{xx} + \alpha_y^2 e_{yy} + \alpha_z^2 e_{zz}) + B_2(\alpha_x \alpha_y e_{xy} + \alpha_x \alpha_z e_{xz} + \alpha_y \alpha_z e_{yz}), \quad (\text{A1})$$

where α_i is the ratio of the i th component of magnetization to the saturation magnetization and the e_{ij} are components of the strain. The linear equations of motion resulting from this energy are given by Kittel.²³ The equations of motion appropriate to parallel pumping are given by Morgenthaler⁶ and Comstock.²² It is seen that for shear waves the coupling constant is B_2 regardless of the direction of propagation. Longitudinal waves are not linearly coupled when the magnetic field is along [100].

For H directed along [111] (z axis), when the y axis is along $[\bar{1}10]$ and the x axis splits two [110] directions (which is a $[11\bar{2}]$ axis), the linear terms in the magnetoelastic energy are^{24,25}

$$U_{\text{me}}^{[111]} = B_2[\alpha_x \alpha_z e_{xz} + \alpha_y \alpha_z e_{yz}] + (B_1 - B_2) \frac{2}{3} \{ \alpha_x \alpha_z [(1/\sqrt{2})(e_{xx} - e_{yy}) + e_{xz}] + \alpha_y \alpha_z [e_{yz} - (1/\sqrt{2})e_{xy}] \}. \quad (\text{A2})$$

For shear strains polarized along H , the linear terms can be written as

$$(2B_1 + B_2)/3 [\alpha_x \alpha_z e_{xz} + \alpha_y \alpha_z e_{yz}]. \quad (\text{A3})$$

Comparing (A3) with (A1) it is seen that the energy with [111] orientation has the same form as for [100] orientation with a mixed combination of coupling constants in the former. Thus, the analysis of Morgenthaler⁶ can be applied to the [111] orientation with $(2B_1 + B_2)/3$ replacing B_2 in all equations.

For shear waves polarized perpendicular to H , the equations of motion resulting from the energy given in (A2) (last term) indicate that the analysis discussed in Ref. 6 is still valid with an effective magnetoelastic coupling constant of $(B_2 - B_1)/(2/3)^{1/2}$. If B_2 is negligible as in lithium ferrite, then the coupling constant is smaller by $1/\sqrt{2}$ than for the case of polarization along H . Since the value of B_1 obtained assuming polarization along H agrees with previously measured values using other methods it seems probable that this was the correct assumption.

It is also observed in case the field is along [111] that linear coupling exists between longitudinal waves propagating in either the x or y direction.

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

²⁴ T. Kasuya (private communication); see Ref. 24.

²⁵ R. C. LeCraw and R. L. Comstock, in *Physical Acoustics*, edited by W. P. Mason (Academic Press Inc., New York, to be published).