MAGNETOELASTIC TUNING OF THE ANTIFERROMAGNETIC RESONANCES IN ORTHOFERRITES

Geoffrey O. White and Carl E. Patton

Abstract - The stress dependences of the two-sublattice antiferromagnetic resonance frequencies in orthoferrites are formulated and estimated for the case of longitudinal stress. With the lower frequency mode, a stress dependence of a few MHz/bar is expected, due primarily to the stress dependence of the in-plane anisotropy. In the temperature region of spontaneous spin rotation, this effect should be much larger. The tuning of the upper mode is expected to be of the order of tens of MHz/bar. This is due largely to the stress dependence of the antisymmetric exchange, and to a lesser degree, to the magnetoelasticity of the out-of-plane anisotropy.

INTRODUCTION

The recent interest in mm wave technology has motivated a search for materials with magnetic resonances at frequencies in excess of 100 GHz. Because the effective field for resonance in an antiferromagnet is the geometric mean of exchange and anisotropy effective fields, virtually any such material with a practically high Néel temperature will satisfy this requirement. By the same token, the magnetoelastic tuning of these resonances might also be large.

The orthoferrites, $RFeO_3$, where R is a rare earth or Y constitute a group of canted antiferromagnets with Néel temperatures in the range 620-740 K [1] that is worthy of consideration. All of these compounds crystalize in the orthorhombically distorted perovskite structure which contains four iron sublattices [2]. The iron spins lie in one of the configurations shown in Fig. 1. All orthoferrites exhibit the Γ_4 Fe spin configuration at high temperatures [3]. At lower temperatures some of the rare earth orthoferrites undergo a continuous spin rotation to the Γ_2 state, or transit to the Γ_o state. With $SmFeO_3$, the transition occurs at about 500 K. In all other cases, the Γ_4 state is maintained down to 100 K or lower.

The iron sublattices have four antiferromagnetic resonance modes, described by Herrmann [4]. The two lower frequency of these are adequately described by a two-sublattice model in which the nearly parallel sublattice spin pairs precess together. Based on the theory, and on low frequency data, these resonances are expected to lie, in zero field, at 300 GHz and up. In fact, they have been observed in $YFeO_3$ at 330 and 520 GHz [5], and in $LaFeO_3$ at 672 and 789 GHz [6]. A notable exception to such high frequencies occurs in the instance of continuous spin rotation, where, at the temperatures bounding the transition, the lowest mode frequency goes to zero [7,8]. The frequencies of the other two modes are much higher, and these modes will not be given further consideration here.

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In this work, the stress dependences of the zero-field antiferromagnetic resonance frequencies of the iron sublattices in orthoferrites under longitudinal applied stress σ are formulated for the two-sublattice model. The magnitudes of these dependences are then estimated, based on available data on magnetoelastic constants of orthoferrites. With the lower frequency xy mode [9], the mechanical tuning of the resonance frequency f_{xy} is primarily due to the variation in anisotropy. For orthoferrites resonating at mm wavelengths, $df_{xy}/d\sigma$ is likely to amount to, at most, a few MHz/bar. In the region of spontaneous spin rotation however, this effect could be much larger, since $df_{xy}/d\sigma$ is proportional to $1/f_{xy}$. With the higher frequency z mode [9], a tuning of tens of MHz/bar is anticipated. This would be due largely to a variation in antisymmetric exchange and, to a lesser degree, to the variation in anisotropy.

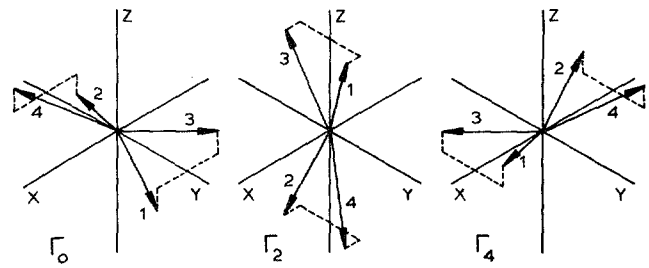

Fig. 1. Iron spin configurations in orthoferrites.

ZERO-STRESS FORMALISM

The antiferromagnetic resonance frequencies can be derived from the expression for the normalized magnetic free energy F/M_o, where F is the free energy and M_o is the sublattice magnetization. For the two-sublattice approximation in zero field, this is given by

$$\begin{aligned}\frac{F}{M_o} &= E\underset{\sim}{m}_1\cdot\underset{\sim}{m}_2 + B_{xx}X_1X_2 + B_{zz}Z_1Z_2 - D(X_1Z_2-X_2Z_1)\\ &- A_{xz}(X_1Z_1-X_2Z_2) - A_{xx}(X_1^2+X_2^2) - A_{zz}(Z_1^2+Z_2^2) \qquad (1)\\ &+ C_{xx}(X_1^4+X_2^4) + C_{zz}(Z_1^4+Z_2^4) - C_{xz}(X_1^2Z_1^2+X_2^2Z_2^2)\\ &+ C_{xxz}(X_1^3Z_1-X_2^3Z_2) + C_{zxz}(Z_1^3X_1-Z_2^3X_2)\ .\end{aligned}$$

In (1), the sublattice vector magnetizations, $\underset{\sim}{M}_1$ and $\underset{\sim}{M}_2$, are defined by

$$\underset{\sim}{m}_i \equiv \frac{\underset{\sim}{M}_i}{M_o} = X_i\hat{x} + Y_i\hat{y} + Z_i\hat{z}\ ,$$

and E is the isotropic exchange field. The terms in B represent the anisotropy due to exchange and/or dipolar interactions [10], D is the antisymmetric exchange field, and the terms in A and C are attributable to crystalline anisotropy. These constants are in field units.

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The method of calculating the resonance frequencies from (1) is well known [9]. To good approximation, considering the relative magnitudes of the effective fields, the frequencies of the two modes are given by

$$(f_{xy}/\gamma_o)^2 = 4EK_{xy} \ , \tag{2a}$$

and

$$(f_z/\gamma_o)^2 = 4E(K_z + \gamma^2 E) \ , \tag{2b}$$

where

$$K_{xy} \equiv A_{xx} - A_{zz} + (B_{xx}-B_{zz})/2 - 2(C_{xx}+C_{xz}/2) \tag{3a}$$

defines the in-plane anisotropy,

$$K_z \equiv A_{xx} + B_{xx}/2 - 2C_{xx} \tag{3b}$$

defines the out-of-plane anisotropy, γ_o is 2.8 MHz/Oe, and $\gamma \sim D/2E$ is the zero field canting angle.

STRESS-DEPENDENT FORMALISM

The strain arising from the application of a mechanical stress to the crystal will alter the antiferromagnetic resonance frequencies. To evaluate such changes in the general case, one would include the strain term in the free energy expression, and then rederive the resonance expressions of (2-3). In the case of longitudinal (non-shear) stress only, shear strains are not generated. Consequently, no new terms are introduced into the free energy expression. The effect of such a stress is merely to alter those terms already present. As the form of the free energy is invariant, so also are the resonance expressions. Thus, the stress dependence of the resonance frequencies, $df/d\sigma$, is obtained simply by differentiating (2) with respect to σ. One obtains

$$\frac{df_{xy}}{d\sigma} = \frac{2\gamma_o^2}{f_{xy}}\left(E\frac{dK_{xy}}{d\sigma} + K_{xy}\frac{dE}{d\sigma}\right) \tag{4a}$$

and

$$\frac{df_z}{d\sigma} = \frac{2\gamma_o^2}{f_z}\left(E\frac{dK_z}{d\sigma} + K_z\frac{dE}{d\sigma} + \frac{D}{2}\frac{dD}{d\sigma}\right). \tag{4b}$$

The stress dependence of an effective field can be related to the magnetostriction constants by expanding the effective field in a Taylor series about the zero-stress strain value, as given by

$$H(c) \equiv H(\underset{\sim}{\varepsilon}_o) + \frac{\partial H}{\partial \varepsilon_i}\bigg|_{\underset{\sim}{\varepsilon}_o} \varepsilon_i + \frac{1}{2}\frac{\partial^2 H}{\partial\varepsilon_i\partial\varepsilon_j}\bigg|_{\underset{\sim}{\varepsilon}_o} \varepsilon_i\varepsilon_j + \ldots \ , \tag{5}$$

where H is the generic effective field, the ε terms comprise the six strain components in the Love notation, ε_o is the strain at zero stress, and summation over repeated indices is implied. The strain derivatives in (5) are referred to as the magnetostriction constants, which will be functions of temperature. The strains are related to the stress through the elasticity tensor. For the case of longitudinal stress in an orthoferrite, this can be written to good approximation as

$$\begin{bmatrix}\varepsilon_1\\ \varepsilon_2\\ \varepsilon_3\end{bmatrix} = \frac{1}{T}\begin{bmatrix}1 & \mu & \mu\\ \mu & 1 & \mu\\ \mu & \mu & 1\end{bmatrix}\begin{bmatrix}X_x\\ Y_y\\ Z_z\end{bmatrix} \ , \tag{6}$$

where the X, Y, and Z are the components of the stress σ in those directions, T is Young's modulus ($\sim 2\times10^{12}$) and μ is Poisson's ratio ($\sim$0.3) [11]. An expression for the stress dependence of the effective field follows from (5-6). For stress along the c axis, for example,

$$H(\sigma) = H(o) + \left[\mu\left(\frac{\partial H}{\mu\varepsilon_1} + \frac{\partial H}{\mu\varepsilon_2}\right) + \frac{\partial H}{\partial\varepsilon_3}\right]\frac{\sigma}{T} + \frac{1}{2}\left[\mu^2\left(\frac{\partial^2 H}{\partial\varepsilon_1^2} + \frac{\partial^2 H}{\partial\varepsilon_2^2} + \frac{\partial^2 H}{\partial\varepsilon_1\varepsilon_2}\right) + \mu\left(\frac{\partial^2 H}{\partial\varepsilon_1\partial\varepsilon_3} + \frac{\partial^2 H}{\partial\varepsilon_2\partial\varepsilon_3}\right) + \frac{\partial^2 H}{\partial\varepsilon_3^2}\right]\frac{\sigma^2}{T^2} \ . \tag{7}$$

The stress derivatives of the effective fields in (4) are obtained by differentiation of (7).

The connection between the stress dependence of the effective fields and the magnetostriction constants is explicitly stated in (7). From this it is apparent that the stress derivatives in (4) involve combinations of the magnetostriction constants and elastic constants in such a way that $dH/d\sigma$ depends upon the direction of the applied stress. For sufficiently large stress, $dH/d\sigma$ will also depend on the magnitude of the stress.

By assigning appropriate values to the terms in (4), the stress dependences of the lower mode frequencies can be estimated. Unfortunately, no complete characterization for the relevant orthoferrites has been reported. Nevertheless, general order-of-magnitude estimates can be fabricated from a composite set of parameters such as those shown in Table I. The zero-stress effective fields in Table I are those of $YFeO_3$, in which zero-field resonance has been observed for both modes [5]. The room temperature values of E and D were estimated from magnetization and low-frequency susceptibility data [12]. The anisotropy values were deduced from (2) based on reported values of 330 and 520 GHz for f_{xy} and f_z respectively (5).

TABLE I

Representative Values of Effective Fields in Orthoferrites, and Estimates of their Stress Dependences at Zero Stress.

Parameter	E	D	K_{xy}	K_z
Zero-Stress Value (Oe)	5.8×10^6	1.3×10^5	600	800
Stress Derivative (Oe/bar)	10	15	0.1	0.1

Few if any of the numerous magnetostriction constants are individually known for any of the orthoferrites. Fortunately, some of their relevant combinations which correspond to the stress derivatives of the effective fields have been measured directly or can be deduced from other measurements. These are estimated, based on data for several orthoferrites, in Table I. Of ocurse, the magnitudes of these quantities will vary somewhat depending upon δ. With the anisotropies this variation should not exceed a factor of three [11].

The stress dependences of the exchange terms can be estimated from the stress dependences of the Néel temperature and the net moment. The Néel temperature is proportional to E. Based upon the pressure dependence of the Néel temperature in several orthoferrites, E can be expected to vary by less than 10 Oe/bar [13]. For this magnitude of $dE/d\sigma$, the stress dependence of the net moment is almost entirely attributable to $dD/d\sigma$. Thus, $dD/d\sigma$ is estimated to be about 15 Oe/bar, based on the reported 20% change in the net moment in $TmFeO_3$ under a c axis compression of 1.5 kbar [11].

The stress derivative of the in-plane anisotropy $dK_{xy}/d\sigma$ in Table I is representative of reported values based on acoustic measurements in $ErFeO_3$ [14,15] and $TmFeO_3$ [11,14]. Because these data were taken in a temperature region near spontaneous spin rotation, this value of $dK_{xy}/d\sigma$ is likely to represent an overestimate in instances where the rare earth ion is not assertive.

An experimental value for $dK_z/d\sigma$ has not been reported, presumably because this requires data on the stress dependence of f_z. Nevertheless, since K_{xy} and K_z are of equivalent magnitude and of the same origin, it is reasonable to expect a value of $dK_z/d\sigma$ roughly equal to that of $dK_{xy}/d\sigma$.

Incorporated into (4a), the data of Table I lead to an expected room-temperature mechanical tuning in f_{xy} of 10 MHz/bar or less. In the vicinity of spontaneous spin rotation, on the other hand, the tuning could become quite large as f_{xy} and K_{xy} become small. In either case, $df_{xy}/d\sigma$ is not appreciably dependent upon $dE/d\sigma$, due to the magnitude of E/K_{xy}.

The stress dependence of f_z is likely to be dominated by the piezomagnetic term, $dD/d\sigma$. This would result in a tuning of tens of MHz/bar. To this may be added a few MHz/bar arising from the $dK_z/d\sigma$ term. As with the xy mode, $dE/d\sigma$ will not contribute significantly to $dK_z/d\sigma$. Because D and $dD/d\sigma$ are essentially constant through spin rotation, the characteristic increase in $df_{xy}/d\sigma$ near spin rotation is not expected for $df_z/d\sigma$.

Data on $dK_{xy}/d\sigma$ indicate that the tuning of f_{xy} will become nonlinear at stresses in excess of about two kilobars [11,14]. In the absence of analogous data on $dD/d\sigma$ and $dK_z/d\sigma$, estimation of the linear tuning range of f_z is not appropriate.

SUMMARY AND IMPLICATIONS

The magnetoelastic tuning of the two lower frequency antiferromagnetic resonance modes of orthoferrites has been considered. For longitudinal stress, the frequency of the lower of these modes may be varied by a few MHz/bar, except in the temperature region of spontaneous spin rotation, where the tuning rate can become very large over a limited stress range. With the higher frequency mode, tuning is likely to be on the order of tens of MHz/bar regardless.

Because of the stress dependences of the resonances discussed here, orthoferrites might conceivably be employed as mechanically tunable mm wave filters. The practicability of such employment however would seem to depend foremost on the magnitude of the tuning relative to the linewidth. Based on very limited information, linewidths of the order of one [16] to tens [17] of GHz may be expected. In terms of the anticipated tunings, this translates into requisite stresses of kilobars or more. Such stresses have been successfully applied in the laboratory to single crystals of hematite [18].

Device implications aside, the stress dependences of the resonance frequencies could be exploited to provide worthwhile information about the materials themselves. Observation of the tuning of the lower mode should yield information regarding $dK_{xy}/d\sigma$. From the stress dependences of the upper mode frequency, and the net moment, a value for $dK_z/d\sigma$ could be obtained. From the stress dependences of the anisotropies may in turn follow information regarding the nature of their origins. For example, the stress dependence of a magnetic dipolar anisotropy should be readily calculable for comparison with experiment.

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