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# Magnetoelastic Interactions in Ferrites

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**Abstract**—Some of the basic sources of magnetoelastic interactions are reviewed. Particular emphasis is given to mechanisms which are appropriate to the ferrosinels and the ferrogarnets. The most important mechanisms involve strain-produced perturbations in the local crystal field which affect the orientation of the orbital magnetic moment through distortions in the electronic charge distribution of the ion. The spin magnetic senses the strain indirectly as a result of the spin-orbit interaction. A fairly precise picture emerges for  $\text{Co}^{2+}$  and  $\text{Fe}^{2+}$  ions in the spinels, since the orbital moments are not "quenched" by the cubic field and the magnetoelastic interactions are calculated as low-order perturbations in the magnetic energy. Since the magnetic moments of most of the trivalent rare earth ions involve an orbital contribution, the rather strong magnetoelastic effects produced by these ions may be rather easily understood, at least qualitatively. The special case of  $\text{Eu}^{3+}$  seems to illustrate the importance of strain-induced variations in the anisotropic exchange coupling. The magnitudes of the magnetoelastic constants of orbitally nondegenerate ions such as

$\text{Fe}^{3+}$ ,  $\text{Ni}^{2+}$ , and  $\text{Mn}^{2+}$  are much more difficult to understand. However, the results of stress-dependent ESR measurements help to illuminate these difficulties. An extensive tabulation of magnetoelastic and magnetostrictive constants is also presented.

## INTRODUCTION

THE SUPPOSITION of some sort of magnetoelastic interaction has played a central role in the understanding of such important magnetic phenomena as magnetostriction, magnetomorphism, domain nucleation, magnon-phonon generation, and magnetic phase transformations. For a cubic material the interaction is commonly treated as a phenomenological contribution to the magnetic free energy of the form

$$F_{ME} = B_1 \left\{ (\alpha_x^2 - \frac{1}{3}) \epsilon_{xx} + (\alpha_y^2 - \frac{1}{3}) \epsilon_{yy} + (\alpha_z^2 - \frac{1}{3}) \epsilon_{zz} \right\} \\ + B_2 \{ \alpha_x \alpha_y \epsilon_{xy} + \alpha_y \alpha_z \epsilon_{yz} + \alpha_z \alpha_x \epsilon_{zx} \} \quad (1)$$

where the  $\alpha$ 's are direction cosines of the magnetization and the  $\epsilon$ 's are the strain components. This form of free energy function leads directly to the familiar two-component expression for the saturation magnetostriction

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TABLE I  
MATERIALS CONTAINING ORBITALLY DEGENERATE MAGNETIC IONS

Material	Temperature (°K)	$\lambda_{100}$ ( $\times 10^6$ )	$\lambda_{111}$	$\lambda_S$	$B_1^*$ ( $\text{cm}^{-1}$ per molecule)	$B_2^*$	Reference
EuIG	4	+110	+20	+56	-650	-110	[30]
EuIG	77	+86	+9.7	+40	-500	-53	[18]
EuIG	300	+21	+1.8	+10	-120	-10	[18]
TbIG	77	+560	+67	+264	-3200	-360	[18]
TbIG	300	+12	-3.3	+3	-69	+18	[18]
YbIG	77	+18.3	-14.4	-1	-110	+8	[31]
Yb <sub>0.1</sub> Y <sub>0.9</sub> IG	4	+7	-6	-1	-41	+34	[32]
Yb <sub>0.1</sub> Y <sub>0.9</sub> IG	77	+1	-4	-2	-6	+24	[32]
CoFe <sub>2</sub> O <sub>4</sub>	77	-830			-490		[14]
CoFe <sub>2</sub> O <sub>4</sub>	300	-450			+260		[14]
Co <sub>0.5</sub> Mn <sub>0.5</sub> Fe <sub>2</sub> O <sub>4</sub>	77	-440			+260		[14]
Co <sub>0.5</sub> Mn <sub>0.5</sub> Fe <sub>2</sub> O <sub>4</sub>	300	-230			+140		[14]
Co <sub>0.3</sub> Mn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub>	300	-200	+65	-40	+120	-58	[33]
Co <sub>0.05</sub> Mn <sub>0.95</sub> Fe <sub>2</sub> O <sub>4</sub>	100	-51			+30		[14]
Co <sub>0.05</sub> Mn <sub>0.95</sub> Fe <sub>2</sub> O <sub>4</sub>	300	-27			+16		[14]
Co <sub>0.05</sub> Ni <sub>0.95</sub> Fe <sub>2</sub> O <sub>4</sub>	100	-75	-50	-60	+44	+44	[34]
Co <sub>0.05</sub> Ni <sub>0.95</sub> Fe <sub>2</sub> O <sub>4</sub>	300	-75	-20	-42	+44	+18	[34]
Co <sub>0.3</sub> Fe <sub>2.2</sub> O <sub>4</sub>	300	-590	+120	-210	+350	-110	[33]
Co <sub>0.3</sub> Zn <sub>0.2</sub> Fe <sub>2.2</sub> O <sub>4</sub>	300	-210	+110	-18	+120	-97	[33]
Co <sub>0.04</sub> Fe <sub>2.96</sub> O <sub>4</sub>	300	-39			+23		[35]
Fe <sub>3</sub> O <sub>4</sub>	124	-23	+55	+24	+13	-48	[36]
Fe <sub>3</sub> O <sub>4</sub>	300	-20	+78	+39	+12	-69	[36]
Fe <sub>0.6</sub> Mn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub>	300	-6	+63	+35	+4	-55	[37]
Fe <sub>0.6</sub> Mn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub>	77	-3	+98	+58	+2	-86	[37]
Fe <sub>0.15</sub> Mn <sub>0.85</sub> Fe <sub>2</sub> O <sub>4</sub>	300	-14	+16	+4	+8	-14	[37]
Fe <sub>0.15</sub> Mn <sub>0.85</sub> Fe <sub>2</sub> O <sub>4</sub>	77	-15	+32	+13	+9	-28	[37]
Fe <sub>0.2</sub> Ni <sub>0.8</sub> Fe <sub>2</sub> O <sub>4</sub>	300	-36	-4	-17	+16	+1	[33]
Fe <sub>0.25</sub> Zn <sub>0.45</sub> Ni <sub>0.3</sub> Fe <sub>2</sub> O <sub>4</sub>	300	-15	+11	+1	+7	-7	[38]
(Fe <sub>2</sub> TiO <sub>4</sub> ) <sub>0.18</sub> (Fe <sub>3</sub> O <sub>4</sub> ) <sub>0.82</sub>	77	+145	+82	+107	-85	-72	[15]
(Fe <sub>2</sub> TiO <sub>4</sub> ) <sub>0.18</sub> (Fe <sub>3</sub> O <sub>4</sub> ) <sub>0.82</sub>	300	+45	+110	+84	-26	-96	[15]
(Fe <sub>2</sub> TiO <sub>4</sub> ) <sub>0.56</sub> (Fe <sub>3</sub> O <sub>4</sub> ) <sub>0.44</sub>	77	+990	+330	+594	-580	-290	[15]
(Fe <sub>2</sub> TiO <sub>4</sub> ) <sub>0.56</sub> (Fe <sub>3</sub> O <sub>4</sub> ) <sub>0.44</sub>	300	+170	+92	+123	-100	-81	[15]
CuFe <sub>2</sub> O <sub>4</sub> (quenched)	300			-16			[39]
CuFe <sub>2</sub> O <sub>4</sub> (annealed)	300			-2			[39]

\*  $B$  values estimated using approximate elastic constants and unit cell sizes.

TABLE II  
MATERIALS CONTAINING ONLY ORBITALLY NONDEGENERATE MAGNETIC IONS

Materials	Temperature (°K)	$\lambda_{100}$ ( $\times 10^6$ )	$\lambda_{111}$	$\lambda_S$	$B_1^*$ ( $\text{cm}^{-1}$ per molecule)	$B_2^*$	Reference
YIG	1.5	-1.1	-5.4	-3.7	+6	+29	[40]
YIG	300	-2.0	-3.6	-3.0	+11	+20	[40]
GdIG	4.2	+7.1	-3.9	~0	-39	+22	[29]
GdIG	77	+4.0	-5.1	-1.5	-23	+29	[31]
GdIG	300	~0	-3.1	-1.9	~0	+17	[31]
NiFe <sub>2</sub> O <sub>4</sub>	4.2	-51.0	-23.7	-34.6	+31	+21	[34]
NiFe <sub>2</sub> O <sub>4</sub>	300	-45.9	-21.6	-31.3	+27	+19	[34]
Li <sub>0.5</sub> Fe <sub>2.5</sub> O <sub>4</sub>	77	-25	-4	-12	+15	+4	[31]
Li <sub>0.5</sub> Fe <sub>2.5</sub> O <sub>4</sub>	300	-31	-3	-14	+19	+3	[31]
Li <sub>0.5</sub> Fe <sub>2.5</sub> O <sub>4</sub>	300	-40			+24	$\ll B_1$	[41]
Li <sub>0.5</sub> Fe <sub>2.5</sub> O <sub>4</sub>	300			-8			[42]
MnFe <sub>2</sub> O <sub>4</sub>	150	-35	+15	-5	+21	-13	[37]
MnFe <sub>2</sub> O <sub>4</sub>	300	-25	+5	-7	+15	-4	[37]
Mn <sub>0.8</sub> Zn <sub>0.1</sub> Fe <sub>2.1</sub> O <sub>4</sub>	300	-14	+14	+3	+8	-12	[33]

\*  $B$  values estimated using approximate elastic constants and unit cell sizes.

with the strains along, respectively, the [100] and [111] directions given by

$$\lambda_{100} = -\frac{2}{3} \frac{B_1}{C_{11} - C_{12}} \quad (2)$$

$$\lambda_{111} = -\frac{1}{3} \frac{B_2}{C_{44}}$$

where the  $C$ 's are cubic elastic constants. More elaborate, multiparameter free energy functions have been developed

to include higher order terms in the  $\alpha$ 's and to account for internal or optical mode strains [1]-[6]. However, these refinements have had little application due to the general scarcity of experimental data on magnetoelastic interactions.

In this talk, we review and suggest some semiquantitative models for the magnitudes of the magnetoelastic constants in ferrosipinel and ferrogarnets. We also present, in Tables I and II, a compilation of representative experi-

mental values of these constants. It is hoped that this data will be of some guidance for workers in the fields of magnetic applications. In preparing this presentation we have drawn heavily on the excellent reviews of this subject by Tsuya [7], Kanamori [8], and Comstock [9]. We treat only the magnetic oxides since the behavior of these materials may be understood in terms of the characteristics of localized electronic states. The description of the magnetoelastic interaction in metals is much too formidable at this time.

Models of magnetoelastic energy are generally related to those of magnetocrystalline anisotropy in that both require a coupling between the direction of the magnetization and the symmetry or changes in symmetry of the lattice. They differ, at least in cubic materials, in the requirement that the former involves interactions quadratic in the  $\alpha$ 's and the latter interactions quartic in the  $\alpha$ 's. The anisotropy models have been more successful since more is known about the relevant crystalline parameters. In magnetoelastic models we must know ultimately the strain variations of some potential—in general a poorly understood set of parameters.

Generally, the potential sources of magnetoelastic interactions may be divided into two separate classes—viz. those involving inter-ionic spin-spin interactions and those involving a sequence of crystal field intra-ionic spin-orbit and inter-ionic spin-spin interactions. Examples of the former are the strain variations in the classical dipole-dipole interaction and in the generalized Heisenberg-Dirac-Van Vleck exchange interaction. The various examples of the latter class treat the strain produced variations in the local electrical crystal fields which, in turn, distort the electronic charge distribution of the ions and, hence, affect the ions' orbital state. The dominantly "spin-only" magnetization then senses indirectly distortions of the crystal field through the spin-orbit interaction.

We first discuss the classical dipole-dipole magnetoelastic interaction which is the only truly "magnetic" interaction and, as such, depends only on the total magnetic moments of the ions disposed on a particular lattice. All of the other mechanisms are basically electrostatic so that their effectiveness is highly sensitive to the orbital character of the ground state of the magnetic ion. Accordingly, we discuss separately, ions with orbitally degenerate and nondegenerate ground states.

#### MAGNETIC DIPOLE-DIPOLE INTERACTION

In principle, the classical dipole-dipole contribution to the magnetoelastic constants should be susceptible to accurate calculation, since the interaction potential and its strain variation are completely determined. Only a lattice sum need be evaluated for each particular crystal structure. It is well known that the dipolar interaction makes no contribution in first-order to the anisotropy of a cubic material. On the other hand, it probably makes a non-negligible contribution to the magnetoelastic energy. There seem to be only two such calculations extant which,

unfortunately, are not presented in any detail. Tsuya [7] finds for nickel ferrite  $B_1 = -10 \text{ cm}^{-1}$  per molecule and  $B_2 = +14 \text{ cm}^{-1}$  per molecule with similar values for other common spinels. Nakamura and Sugiura [10] find for YIG  $B_1 = -3.3 \text{ cm}^{-1}$  per molecule and  $B_2 = +2.2 \text{ cm}^{-1}$  per molecule. We see from a comparison with the experimental values for these compounds in Table II that the dipolar mechanism accounts for some appreciable fraction, but not all, of the observed constants. However, it is in these *weak* magnetoelastic substances that the dipolar interaction plays its most important role. It is useful to note that  $(B_1/B_2)_{a-d} = -\frac{3}{2}$  for any cubic lattice. We see that none of the materials listed in Tables I and II conform to this ratio. We conclude that the dipolar interaction may make some contribution to the magnetoelastic constants of cubic oxides, but certainly not a dominant one. In noncubic substances the dipolar interaction may be of greater importance.

#### ORBITALLY DEGENERATE MAGNETIC IONS

The largest magnetoelastic constants arise in those materials which contain ions having appreciable orbital degeneracy in the ground state configuration. The outstanding examples of this situation are  $\text{Co}^{2+}$  and  $\text{Fe}^{2+}$  ions in the ferrosinels and all of the trivalent rare earth ions (except  $\text{La}^{3+}$ ,  $\text{Gd}^{3+}$ , and  $\text{Lu}^{3+}$ ) in the ferrogarnets. In these materials, magnetoelastic effects are calculated, essentially, as low-order perturbations in the magnetic energy and thus we might expect fairly reliable results.

#### Divalent Cobalt Ion

Slonczewski [11]–[13] has carried out the most complete calculation of this sort in treating divalent cobalt ion substitutions in  $\text{MnFe}_2\text{O}_4$  and  $\text{Fe}_3\text{O}_4$ . In the ferrosinels the cobalt ion occupying the octahedral site has an orbital triplet lowest arising from the predominantly cubic, local crystal field of the oxygen anions. This orbital degeneracy is the first prerequisite in accounting for the large cobalt magnetoelastic constants listed in Table I. However, ions on the octahedral site are also subjected to a weaker trigonal crystal field which arises, mainly, from cations dispersed on neighboring octahedral sites. The trigonal field partially lifts the degeneracy of the triplet, but still leaves an orbital doublet lowest. As illustrated in Fig. 1, the further sequential application of inter-ionic spin-spin exchange and intra-ionic spin-orbit interactions completely removes the spin and orbital degeneracy of the ion. Any strain imposed on the crystal will introduce a new crystal field component  $\delta V$  of lower symmetry. The magnetoelastic energy associated with a given strain may be calculated as perturbations on the ground state orbital doublet. Two types of terms are of importance. In first-order,  $\delta V$  may lift the degeneracy of the doublet and in second-order, interactions with the excited orbital singlet may introduce a term of order  $(\lambda/\Delta)\delta V$ . Slonczewski's model, at least quantitatively, explains the relatively smaller magnetoelastic effects of cobalt ions in  $\text{MnFe}_2\text{O}_4$

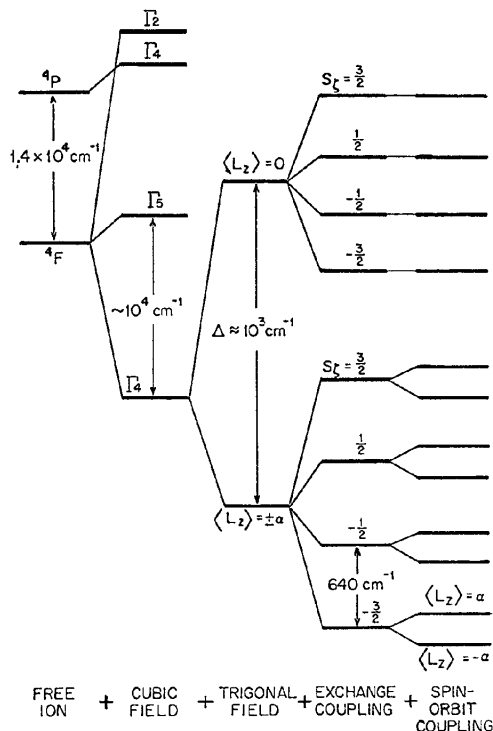


Fig. 1. Energy levels of divalent cobalt ion in ordered ferros spinels (after Slonczewski).

and  $\text{NiFe}_2\text{O}_4$  in contrast to the very much stronger effects in  $\text{Fe}_3\text{O}_4$ . In the former compounds, the random distribution of ions on the neighboring octahedral sites reduces the trigonal splitting and introduces crystal fields of lower symmetry which lift the degeneracy of the orbital doublet. A diminished strain dependent energy follows from this reduction in orbital degeneracy. However, in magnetite, i.e.,  $\text{Fe}_3\text{O}_4$ , at temperatures above the cubic-orthorhombic transition, electron "hopping" events between divalent and trivalent iron ions tend to average out nontrigonal crystal field components and the strain dependent energy is relatively larger. One of the interesting characteristics of this model is the prediction of a *saturation* in the temperature dependence of the magnetoelastic constants at temperatures below a temperature characteristic of the splitting between the ground and first excited states of the cobalt—typically 50–100°K. Such saturation behavior is clearly exhibited by all cobalt containing ferros spinels.

Slonczewski's model is "single ion" in the sense that only the local interactions of the cobalt ion are considered and interactions between cobalts are neglected. The validity of his model is experimentally justified by the observation that the magnetostriction constants of the compounds  $\text{Mn}_{1-x}\text{Co}_x\text{Fe}_2\text{O}_4$  are strictly linear in cobalt concentration over the entire compositional range  $0 \leq x \leq 1$ , [14].

#### Divalent Iron Ion

In most cases noted in Table I the contribution of ferrous ions to the magnetoelastic constants of ferro-

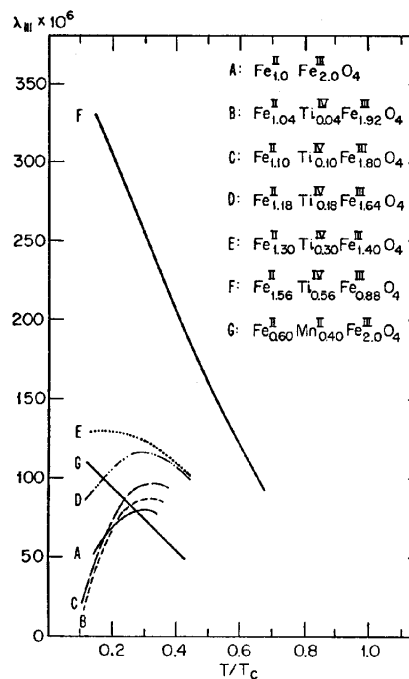


Fig. 2. A comparison of temperature dependence of the ferrous ion contribution to the magnetostriction constant  $\lambda_{111}$  in a number of ferros spinels (see Table I for references).

spinels is, at least, an order of magnitude less than the comparable effect of divalent cobalt ions. Such a reduction may be easily understood in terms of the picture presented above. The  $\text{Fe}^{2+}$  ions again preferentially occupy the octahedral site and have an orbital triplet lowest in a cubic field. However, in this case, for the same sign of the trigonal crystal field the orbital singlet is lowest in energy [11]. The ion only senses strain induced potentials by virtue of the small admixture of excited doublet states into the ground singlet. The admixing of these states is produced by the spin-orbit interaction. The magnetoelastic energy comes in as a perturbation of order  $(\lambda/\Delta)^2 \delta V$  which is a factor  $(\lambda/\Delta)$ —generally of order 1/10—down from the cobalt values.

The measurements of Syono [15] on the spinel titanomagnetites pose some interesting problems in the interpretation of the magnetoelastic interaction of ferrous ion. Some of Syono's data on the system  $(\text{Fe}_2\text{TiO}_4)_x(\text{Fe}_3\text{O}_4)_{1-x}$  is plotted in Fig. 2. For compositions near magnetite, i.e.,  $x < 0.2$ , the magnetostriction *decreases* at low temperatures in striking contrast with the behavior of most other magnetic systems. Perhaps an explanation of this anomalous behavior lies in the increasing range of ionic ordering in this material as the temperature decreases [16]. Such an ordering effect might tend to increase  $\Delta$  and diminish the magnetoelastic constants. However, the most surprising aspect of Syono's data is the enormous increase in magnetostriction values at large ferrous ion concentrations. Values comparable to, or greater than, those of cobalt are observed. It has been conjectured [15] that this strong effect is associated with ferrous ions on the tetrahedral site where the orbital doublet is lowest

in a cubic field. This seems like a rather unreasonable explanation of the large values since the degeneracy of the doublet is completely removed by possible lower symmetry fields arising, perhaps, from Jahn-Teller distortions. In fact, divalent copper on the octahedral site, which also has an orbital doublet lowest, contributes little to the magnetostriction [7], see  $\text{CuFe}_2\text{O}_4$  (annealed) value in Table I. A slightly more attractive explanation for the augmented magnetoelastic interaction might lie in the supposition of a reversal in the sign of the trigonal field on the octahedral site. The possibility of such a reversal in sign has been made reasonable by the calculation of Smit et al. [17] on "normal" spinels. With a reversal in sign  $\text{Fe}^{2+}$ , appearing like  $\text{Co}^{2+}$ , now has a magnetoelastic energy of order  $(\lambda/\Delta)\delta V$ .

### Trivalent Rare Earth Ions

In treating the magnetoelastic properties of rare earth ions, we must be guided by rather different considerations, since the spin-orbit interaction dominates all crystal field influences on these ions. The magnetic states are characterized by the total angular momentum  $J$  and, perforce, sense perturbations due to crystal fields directly through the orbital contribution to  $J$ . As the values in Table I suggest, all of the non- $S$ -state rare earth ions might be expected to exhibit large magnetoelastic constants. We examine here the characteristics of three particular ions—viz.  $\text{Tb}^{3+}$ ,  $\text{Yb}^{3+}$ , and  $\text{Eu}^{3+}$ —in the ferrogarnets.

The measurements of Iida [18], quoted in Table I, show that  $\text{TbIG}$  has extremely large magnetostriction values at low temperatures. Calculations of the energy levels of  $\text{Tb}^{3+}$  in the crystal field of the rare earth, dodecahedral site in the garnets show that this non-Kramer's ion has many closely spread low lying levels [19], [20]. The rapid variation of these levels with the orientation of net magnetization produce "near-crossing" in energy levels which account for many of the curious properties of ferromagnetic resonance in this material [19]. The character of the energy levels and, in particular, the position of "near-crossings" is highly sensitive to the value of the rhombic component of the local field [20]. In view of this sensitivity to fields of low symmetry, the enormous size of the linear magnetostriction constants seems quite reasonable.

In contrast, the magnetostriction constants of  $\text{YbIG}$  are of an order of magnitude less than those of  $\text{TbIG}$ . The  $J = 7/2$  ground state of the  $\text{Yb}^{3+}$  ion is split by the cubic field of the dodecahedral site leaving a well separated Kramer's doublet lowest. It has been well established that the magnetic properties of the  $\text{Yb}^{3+}$  in ferrogarnets are determined by an anisotropic splitting  $\Delta$  of this doublet. Wickersheim [21] has determined  $\Delta$  optically as a function of the orientation of the magnetization. The anisotropy in the splitting is determined mainly by the anisotropy in the rare earth-ferric ion exchange potential [22] and depends only weakly on crystal fields of lower than cubic symmetry at the rare earth site. It may be

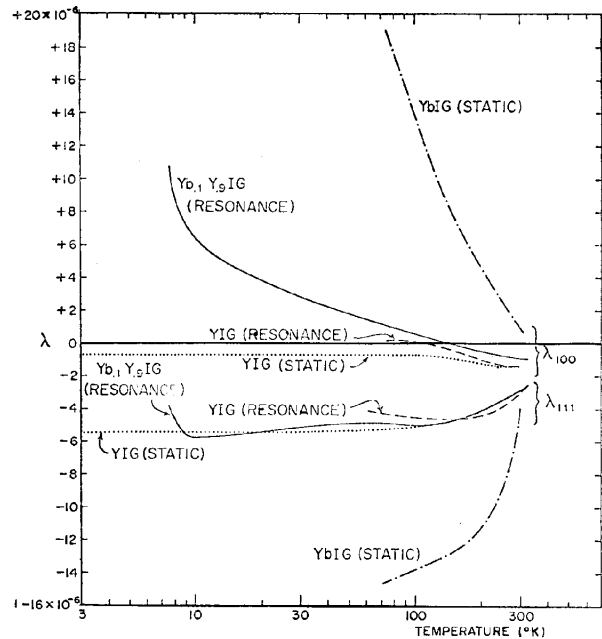


Fig. 3. The temperature dependence of magnetostriction constants of YIG and  $\text{Yb}^{3+}$  doped YIG (see Tables I and II for references).

shown quite simply that the magnetoelastic constants of  $\text{Yb}^{3+}$  ion should be of the order  $\Delta(\partial \ln \Delta/\partial \epsilon)$ . Given  $\Delta \sim 20 \text{ cm}^{-1}$  and  $|\partial \ln \Delta/\partial \epsilon| \sim 5$ , we may easily account for the magnitude of the low temperature values listed for  $\text{YbIG}$  in Table I. It might be expected that the magnetoelastic constant would saturate at temperatures below  $\Delta/k$  when the upper member of the doublet becomes depopulated. A suggestion of this saturation is seen in Fig. 3 for the data above  $10^\circ\text{K}$ . Below  $10^\circ\text{K}$  there is a rapid rise in the magnetostriction which is probably due to the influence of small amounts of  $\text{Yb}^{3+}$  ion on the octahedral sites of the garnet [23]. For these sites the sign of the cubic field is revised and lower symmetry fields are of crucial importance in determining the ordering of the lowest energy levels.  $\text{Yb}^{3+}$  on the octahedral sites becomes akin to  $\text{Tb}^{3+}$  on the dodecahedral sites. These "special," wrongly placed ions lead to the various anomalies in the ferromagnetic resonance of  $\text{Yb}^{3+}$  doped YIG [24].

An interesting special case of  $\text{EuIG}$  demonstrates the importance of the strain dependence of anisotropic exchange interactions as a source of magnetoelastic energy. The ground state of  $\text{Eu}^{3+}$  has  $J = 0$  and is, of course, nonmagnetic. However, the  $\text{Eu}^{3+}$  ion has a *Van Vleck* moment as a result of the exchange interaction which, in second order, mixes some of the excited  $J = 1$  state into the ground state [25]. The details of the crystal field seem to have a negligible effect on this moment. Thus, the appreciable anisotropy of  $\text{EuIG}$  must then arise from anisotropy in the exchange interaction. Roughly speaking, we then take

$$(B(0^\circ\text{K}))_{\text{EuIG}} = 2 \chi H_{ez}^2 \frac{\partial \ln H_{ez}}{\partial \epsilon} \quad (3)$$

Again a value  $2 \chi H_{ez}^2 \sim 100 \text{ cm}^{-1}$  and  $|\partial \ln H_{ez}/\partial \epsilon| \sim 5$  yields a reasonable order of magnitude agreement with the values in Table I.

## ORBITALLY NONDEGENERATE MAGNETIC IONS

The source of the magnetoelastic interactions for ions which have orbitally nondegenerate ground states is a most difficult theoretical question. These ions may be of two different sorts: 1) ions which have a free ion  $S$ -state configuration, e.g.,  $Mn^{2+}$ ,  $Fe^{3+}$ , and  $Gd^{3+}$ ; 2) quenched ions which have an orbital singlet lowest under the influence of a cubic crystal field, e.g.,  $Ni^{2+}$  in the spinels. Since there is no interaction between the spherically symmetric charge distribution of the ground state of these ions and the local crystalline potential, we might, from the simplest point of view, accept very weak magnetoelastic interactions arising solely from the dipolar mechanism in materials containing these ions. However, as the data in Table II demonstrates, the orbital singlet ions have appreciable magnetoelastic interactions. Of course, it is well known from ESR work that the crystal fields do indeed influence the magnetic properties of these ions. This influence comes about due to the admixture of higher orbital states into the ground state via the spin-orbit interaction or possibly through intra-ion spin-spin interactions. To understate the issue, it is difficult to calculate these effects. One of the principal problems in these calculations lies in the sensitivity of the results to the values of energy splittings between the ground and excited orbital states. These splittings are not known accurately in general.

Fortunately, we may take another tack in assessing the effectiveness of crystal interactions in producing magnetoelastic effects. If the ESR spectrum of a magnetically dilute ion is observed under conditions of uniaxially stress, the shifts in the spectrum may be related to an additional term in the ion's spin Hamiltonian of the form

$$\mathcal{H}_{ME} = \sum_{ij} G_{ij} \epsilon_{ij} S_i S_j \quad (4)$$

where the indexes specify the coordinate axis of the crystals. For a cubic environment this term reduces to

$$\mathcal{H}_{ME} = G_{11} \{ [\epsilon_{xx} - \frac{1}{2}(\epsilon_{yy} + \epsilon_{zz})] S_x^2 + \dots \} \\ + G_{44} \{ \epsilon_{xy} [S_x S_y + S_y S_x] + \dots \}. \quad (5)$$

If the  $G$  parameters of a given ion in a particular structure are known, then we might obtain a good approximation to the low temperature, magnetoelastic constants of the corresponding magnetically ordered substance. At low temperatures,  $F_{ME} \sim \langle S | \mathcal{H}_{ME} | S \rangle$  and

$$B_1 = \frac{3}{2} S(S - \frac{1}{2}) G_{11} \quad (6) \\ B_2 = 2S(S - \frac{1}{2}) G_{44}.$$

Unfortunately, careful ESR measurements of this sort have only been carried out on one crystal structure, viz. cubic MgO [26, 27, 28]. However, even this limited amount of data provides us with some useful guidance. The cation site in MgO has an octahedral field of strictly cubic symmetry. The data on  $Fe^{3+}$  and  $Mn^{2+}$  does not agree with theory to any degree of precision. As mentioned above,

TABLE III  
MAGNETOELASTIC CONSTANTS IN CUBIC MgO

Ion	$G_{11}$ ( $cm^{-1}$ per unit strain)	$G_{44}$	$B_1$ ( $cm^{-1}$ per ion)	$B_2$
$Mn^{2+}$	+1.50	-0.315	+11	-3
$Fe^{3+}$	+5.49	-0.825	+41	-8
$Ni^{2+}$	+57	+36	+43	+36

such a disagreement is not surprising in view of the high-order perturbation involved in the calculations. Some of the ESR data and the deduced magnetoelastic constants are presented in Table III. Even though these results are only vaguely applicable to either ferrosinels or garnets we must conclude, contrary to Tsuya's speculations [7], that the crystal field effects lead to important contributions in the magnetoelastic constants of orbital singlets. The relative ordering of the strength of interaction in Table III, i.e.,  $B(Ni^{2+}) > B(Fe^{3+}) > B(Mn^{2+})$  is in substantial agreement with the general trend of the data quoted in Table II.

Recently, a direct stress dependent measurement of the ESR of  $Gd^{3+}$  in a diamagnetic garnet host has been used in a similar way to evaluate the magnetoelastic constants of GdIG [29].

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