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*Present address: Physics Department, Purdue University, Lafayette, Indiana.

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MAGNETOSTRICTION CONSTANTS OF THE RARE-EARTH GARNETS FROM THE PRESSURE DEPENDENCE OF ELECTRON-PARAMAGNETIC-RESONANCE SPECTRA*

T. G. Phillips† and R. L. White

Stanford Electronics Laboratories, Stanford University, Stanford, California

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We wish to report a technique for determining the single-ion contribution to the magnetoelastic constants of magnetic insulators, with particular reference to the contribution of Gd^{3+} to the magnetostriction constants of gadolinium iron garnet (GdIG). We proceed by relating the macroscopic magnetoelastic constants of the crystal to the microscopic magnetoelastic constants of the constituent ions, and then determining the relevant microscopic constants from the variation of the EPR spectra of the ions under uniaxial pressure.

The linear magnetostriction of ferromagnetic or ferrimagnetic crystals arises from the strain dependence of the magnetic anisotropy. For a cubic crystal, the magnetoelastic energy is usually written[1]

$$E_{\mathrm{ME}} = \sum_{ijkl} F_{ijkl}^{\mathrm{cubic}} \epsilon_{ij}\alpha_k\alpha_l$$

$$= B_1[(\alpha_1^2 - \tfrac{1}{3})\epsilon_{xx} + (\alpha_2^2 - \tfrac{1}{3})\epsilon_{yy} + (\alpha_3^2 - \tfrac{1}{3})\epsilon_{zz}]$$

$$+ B_2[\alpha_1\alpha_2\epsilon_{xy} + \alpha_2\alpha_3\epsilon_{yz} + \alpha_3\alpha_1\epsilon_{zx}]. \quad (1)$$

We note first that the cubic magnetoelastic tensor $F_{ijkl}^{\mathrm{cubic}}$ has only two independent constants, B_1 and B_2, closely related to the magnetostriction constants λ_{100} and λ_{111}. We note secondly that the magnetoelastic tensor involves terms quadratic in the spin coordinates (the direction cosines of the magnetization) rather than biquadratic in the spin coordinates as is the case for the magnetic anisotropy. It therefore follows that effects involving energies quadratic in the spin coordinates (required by symmetry to cancel out in first order in the total anisotropy energy) will dominate the magnetoelastic energy, and that the success of the single-ion model of magnetic anisotropy[2-4] in the magnetic insulators does not imply the success of a single-ion model of magnetoelastic effects.

In the (cubic) rare-earth garnets, the rare-earth ions occupy six magnetically inequivalent sites each of orthorhombic symmetry. For the S-state ion Gd^{3+}, we write the magnetic Hamiltonian

$$\mathcal{H}_M = \sum_\sigma g\beta\vec{S}^\sigma\cdot\vec{H}_{\mathrm{eff}} + \sum_\sigma \vec{S}^\sigma\cdot D^\sigma\cdot\vec{S}^\sigma + \text{terms in } S^4, S^6$$

$$+ \text{terms involving more than one ion}, \quad (2)$$

where the summation in σ is over the six inequivalent sites. For the iron garnets, $\vec{H}_{\mathrm{eff}}$ will be the molecular field arising from the

interaction of the rare-earth ions with its iron neighbors (assumed isotropic for the S-state ions Fe^{3+} and Gd^{3+}); for the diamagnetic garnets, $\vec{H}_{eff}$ will be the applied external field. The terms in $\vec{S}^{\sigma}\cdot D^{\sigma}\cdot\vec{S}^{\sigma}$ are single-ion crystal-field energy terms quadratic in the spin coordinates and responsible, therefore, for the magnetoelastic energy of Eq. (1). The tensor D^{σ} and its strain dependence $\partial D^{\sigma}/\partial\epsilon_{ij}$ can be determined from the paramagnetic resonance spectrum of Gd^{3+} dilutely incorporated into the diamagnetic garnet $Y_3Ga_5O_{12}$ and subjected to uniaxial strain.

We now assume that only single-ion effects are important in the magnetoelastic energy, and test this hypothesis ultimately by comparing our deductions with experimental data. With this assumption we may write the contribution to the magnetoelastic energy of the crystal of one rare-earth ion on site σ, subjected to a field H_0 in some given direction, as

$$E_{ME}=\sum_{ijkl}\left(\frac{\partial\mathcal{H}_M^{\sigma}}{\partial\epsilon_{ij}}\right)_0\epsilon_{ij}=S^2\sum_{ijkl}\left(\frac{\partial D^{\sigma}}{\partial\epsilon_{ij}}\right)_0\epsilon_{ij}\alpha_k\alpha_l$$

$$=S^2\sum_{ijkl}F_{ijkl}{}^{\sigma}\epsilon_{ij}\alpha_k\alpha_l. \qquad (3)$$

The microscopic magnetoelastic tensor $F_{ijkl}{}^{\sigma}$ of orthorhombic symmetry appropriate to the rare-earth site has twelve components, of which nine are independent.[5] Though these can, in principle, all be obtained from the strain-dependent EPR spectra, we can avoid this task by observing that, for the single-ion model, $F_{ijkl}{}^{\text{cubic}}=\sum_{\sigma}F_{ijkl}{}^{\sigma}$; i.e., the simpler cubic macroscopic tensor must be obtainable by a summation over the sites of the microscopic tensors. If one applies to the crystal a pressure $\vec{P}$ and a field $\vec{H}_0$ in some arbitrary directions and measures the change in crystal-field energy, $S^2\delta D^{\sigma}(\vec{H}_0,\vec{P})$, of each site, it is possible to obtain, by summing over the δD^{σ}, the appropriate macroscopic magnetoelastic constant $F_{ijkl}{}^{\text{cubic}}$. In particular, if one applies a uniaxial pressure $\vec{P}$ along the $\langle 100\rangle$ direction,

$$\frac{N}{6}S^2\sum_{\sigma}\delta D^{\sigma}=\frac{2}{3}\frac{B_1}{C_{11}-C_{12}}P=-\lambda_{100}P \quad (H_0\parallel P),$$

$$=-\frac{2}{6}\frac{B_1}{C_{11}-C_{12}}P=\frac{\lambda_{100}}{2}P \quad (H_0\perp P), \qquad (4)$$

and similarly for $\langle 111\rangle$. N is the number of rare-earth ions per cc, and six enters the equation because of the six inequivalent sites. The C's are the appropriate elastic constants.

Proceeding in this fashion we obtain the single-ion contribution of the Gd^{3+} ions to the 0°K magnetostriction constants of GdIG to be

$$\lambda_{100}=(+7.3\pm 2)\times 10^{-6},$$

$$\lambda_{111}=(+5.0\pm 2)\times 10^{-6}.$$

The latest results of Clark and DeSavage give,[6] at 4.2°K,

$$\lambda_{100}(\text{GdIG})-\lambda_{100}(\text{YIG})=(+8.2\pm 2)\times 10^{-6},$$

$$\lambda_{111}(\text{GdIG})-\lambda_{111}(\text{YIG})=(+1.5\pm 2)\times 10^{-6}.$$

The agreement is sufficiently good, especially for the larger λ_{100}, to conclude that the single-ion model is appropriate to Gd^{3+} in the rare-earth garnet, and that one can deduce macroscopic magnetoelastic constants from measurements of more microscopic parameters such as the strain dependence of the crystal field.

These experiments were carried out at 9 kMc/sec and at 78°K. The major experimental difficulty is site identification in the resonance experiment, and this was aided by previous work.[4] The pressures used were of the order of 5×10^3 kg/cm^2, and the largest observed line shifts of the order of 200 Oe.

We hope to extend these experiments shortly to other rare-earth ions in the garnet structure.

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†Permanent address: Clarendon Laboratory, Oxford University, Oxford, England.

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