VI. CONCLUSIONS

The large field-induced reflectance modulation in ferroelectrics appears to be associated, at least in part, with band structure changes due to ionic displacements within the elementary cell. Because of its relationship with the electro-optic effect, usually observed in transmission, the ER stands as a complementary technique to determine the electro-optic coefficients in the regions where the material is not transparent. The ER spectra of $KTaO_3$, $KTaO_3$-$KNbO_3$, and $BaTiO_3$ present a number of singularities which have been tentatively assigned to band-structure critical points. The experimental results suggest that more theoretical knowledge of the band structure and of its changes with lattice polarization is of great necessity.

ACKNOWLEDGMENTS

The authors are very much indebted to D. Kahng for many profitable suggestions and continuous encouragement. Thanks are due to J. R. Brews for his valuable assistance in the application of symmetry arguments and to E. O. Kane for a critical reading of the manuscript. In many instances, the authors have benefited from discussions with Y. S. Chen and C. N. Berglund and from the technical assistance of W. J. Sundburg, V. J. DeLucca, and M. V. DePaolis.

PHYSICAL REVIEW VOLUME 153, NUMBER 2 10 JANUARY 1967

Single-Ion Magnetostriction in the Iron Group Monoxides from the Strain Dependence of Electron-Paramagnetic-Resonance Spectra*

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(Received 14 July 1966)

The single-ion contribution to magnetostriction in the transition metal monoxides NiO, MnO, FeO, and CoO is calculated on a very simple model and found to account for substantially all the observed magnetostriction. Exchange is included in a molecular-field approximation, and the single-ion magnetoelastic parameters $F_{ijkl}=(\partial g_{kl}/\partial\epsilon_{ij})_0$ and $G_{ijkl}=(\partial D_{kl}/\partial\epsilon_{ij})_0$ are obtained from data on the electron paramagnetic resonance of Ni^{2+}, Mn^{2+}, Fe^{2+}, and Co^{2+} in MgO subject to uniaxial stress. Most of these data are available from the existing literature.

I. INTRODUCTION

THE linear magnetoelastic properties of magnetic materials all have their physical origin in the strain dependence of the magnetic anisotropy energy of the material. The magnetic anisotropy energy may in turn have its origin in single-ion effects which connect the spin direction of a given ion with its local crystallographic environment, or in multi-ion effects such as anisotropic exchange or dipole-dipole interactions which connect the orientation of the spins to the spatial characteristics of the spin array. We have recently shown[1] that in certain of the rare-earth iron garnets the single-ion effects are dominant in the determination of the magnetoelastic constants (specifically the magnetostriction constants) and that, further, the strain dependence of the single-ion parameters may be conveniently determined from the pressure dependence of the electron-paramagnetic-resonance spectrum of the relevant ions. We wish now to extend these ideas and arguments to the transition metal monoxides, and in particular to examine the magnetostriction constants of NiO, MnO, FeO, and CoO.

II. SPIN CONFIGURATIONS AND CRYSTALLOGRAPHIC DISTORTIONS IN THE TRANSITION METAL MONOXIDES

The transition metal monoxides NiO, MnO, FeO, and CoO all crystallize in the cubic NaCl structure and have cubic symmetry above their magnetic ordering (Néel) temperatures. Below the Néel temperature a slight distortion from cubic symmetry occurs in each case.[2–6] The magnetic order in all cases is such that the spins are ferromagnetically aligned within (111) sheets, with successive sheets antiferromagnetically aligned.[7–11] The

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* Work supported in part by the Advanced Research Projects Agency through the Center for Materials Research at Stanford University and in part by the National Science Foundation through NSF Grant No. GK-202.

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TABLE I. A summary of crystal distortions and spin configurations in antiferromagnetic iron group monoxides.

Compound	Distortion	Spin direction	T_N (°K)
NiO	approx. trigonal, $\alpha<60°$ (contraction along [111])	in ferromagnetic (111) plane	520
MnO	trigonal, $\alpha<60°$ (contraction along [111])	in ferromagnetic (111) plane	122
FeO	trigonal, $\alpha>60°$ (expansion along [111])	⊥ ferro (111) plane ‖ [111] expansion axis	198
CoO	approx. tetragonal $c/a<1$	approx. ‖ [001]	291

spin directions and distortion directions for the various transition monoxides are summarized in Table I.

The NaCl structure with ferromagnetically aligned (111) sheets has the property that a contraction along the (111) direction perpendicular to the ferromagnetic sheets will decrease the spacing between antiferromagnetically aligned nearest-neighbor cations more rapidly than the spacings between ferromagnetically aligned next-nearest-neighbor cations.[12] Such a contraction will generally be favorable energetically, and may be expected to occur. The resultant trigonal contraction is, on the approximation of isotropic exchange, independent of the crystallographic orientation of the spins and therefore uncorrelated with spin direction. It is called exchange striction. The magnetoelastic distortions which *are* correlated with spin orientation, and with which we are primarily concerned here, will be mounted on top of the exchange striction and may be either larger or smaller than the exchange striction.

III. MAGNETOSTRICTION AND THE SINGLE-ION HAMILTONIAN

In cubic ferro-, ferri-, and antiferromagnetic materials, magnetostriction can be described by the relation

$$\frac{\delta l}{l}=\tfrac{3}{2}\lambda_{100}(\alpha_1^2\beta_1^2+\alpha_2^2\beta_2^2+\alpha_3^2\beta_3^2-\tfrac{1}{3})+3\lambda_{111}(\alpha_1\alpha_2\beta_1\beta_2+\alpha_2\alpha_3\beta_2\beta_3+\alpha_3\alpha_1\beta_3\beta_1),\quad(1)$$

where the α's and β's are the direction cosines of the magnetization and the strain, respectively, with respect to the cubic crystal axes. In ferromagnetic or ferrimagnetic materials λ_{100} and λ_{111} can be measured by varying the direction of M through the influence of an external magnetic field and observing the change in sample dimensions. For an antiferromagnetic material the direction of the sublattice M_i is much less responsive to external fields, and one has primarily data on the deformation along or perpendicular to some particular spin direction fixed by the crystalline magnetic anisotropy as the crystal orders magnetically. We propose to interpret such data on the transition metal monoxides.

The magnetoelastic energy of the cubic crystal may be written[13]

$$E_{\mathrm{ME}}=B_1[(\alpha_1^2-\tfrac{1}{3})\epsilon_{xx}+(\alpha_2^2-\tfrac{1}{3})\epsilon_{yy}+(\alpha_3^2-\tfrac{1}{3})\epsilon_{zz}]+B_2[\alpha_1\alpha_2\epsilon_{xy}+\alpha_2\alpha_3\epsilon_{yz}+\alpha_3\alpha_1\epsilon_{zx}],\quad(2)$$

where the α_i are again the direction cosines of the magnetization, the ϵ_{ij} are the strains in Love's notation,[14] and B_1 and B_2 are called the first and second magnetoelastic constants of the material. B_1 and B_2 are related to λ_{100} and λ_{111} by the expressions

$$\lambda_{100}=-\frac{2}{3}\frac{B_1}{c_{11}-c_{12}},\quad \lambda_{111}=-\frac{B_2}{3c_{44}},\quad(3)$$

where the c_{ij} are the cubic elastic constants in the Voigt notation.

As remarked in the opening sentence of our Introduction, the magnetoelastic properties of magnetic materials are rooted in the strain dependence of the magnetic anisotropy energy. Let us therefore expand the anisotropy energy in the lattice strains about the equilibrium configuration

$$E_{\mathrm{A}}=(E_{\mathrm{A}})_0+\sum_{ij}\left(\frac{\partial E_{\mathrm{A}}}{\partial\epsilon_{ij}}\right)_0\epsilon_{ij}+\cdots.\quad(4)$$

We see that the terms in the magnetoelastic energy linear in the strains [as in Eq. (2)] are given by

$$E_{\mathrm{ME}}=\sum_{ij}\left(\frac{\partial E_{\mathrm{A}}}{\partial\epsilon_{ij}}\right)_0\epsilon_{ij}.\quad(5)$$

The anisotropy energy E_{A} is an explicit function of the direction cosines of the magnetization; from Ref. 1 or by inspection of Eq. (2) we see that the relevant terms in the magnetoelastic energy are quadratic in the direction cosines of the magnetization. We therefore write

$$E_{\mathrm{ME}}=\sum_{ijkl}\left(\frac{\partial E_{\mathrm{A}}(k,l)}{\partial\epsilon_{ij}}\right)_0\epsilon_{ij}\alpha_k\alpha_l=\sum_{ijkl}B^0{}_{ijkl}\epsilon_{ij}\alpha_k\alpha_l.\quad(6)$$

The magnetoelastic tensor $B^0{}_{ijkl}$ of a cubic crystal may have two or three independent elements depending upon the microscopic origin of the anisotropy energy. For the case of three independent constants the analysis of Kittel[13] must be slightly generalized and leads to the results (in Voigt notation)

$$\lambda_{100}=-\frac{2}{3}\frac{(B^0{}_{11}-B^0{}_{12})}{c_{11}-c_{12}},\quad \lambda_{111}=-\frac{2B^0{}_{44}}{3c_{44}}.\quad(7)$$

We may make the connection between the formal phenomenological description of magnetoelasticity above

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and the microscopic mechanisms underlying the phenomenon by way of the Hamiltonian of the magnetic ions. We shall adopt a single-ion point of view and show that the principal features of magnetostriction in the transition metal monoxides (exclusive of the exchange striction) can be explained within this framework. Consistent with this point of view we shall simulate exchange (a cooperative phenomenon) with a molecular field. In this approximation the single-ion Hamiltonian

$$\mathcal{H}_{\mathrm{A}}=\beta\mathbf{H}_{\mathrm{eff}}\cdot g\cdot\mathbf{S}+\mathbf{S}\cdot D\cdot\mathbf{S}. \qquad (8)$$

Higher order terms in S are neglected, since we are concerned only with the magnetoelastic effects quadratic in the spin coordinates. The single-ion spin Hamiltonian applies, as usual, to some low-lying manifold of states, and within this manifold the effect of the molecular field is sufficiently great that the first term of the spin Hamiltonian dominates and we are in the Zeeman quantization limit. In the ordered magnetic state the ions are primarily in an extremal $S_z=-S$ state, so we may derive the magnetoelastic tensor of the ordered material from the strain dependence of this extremal single-ion state.

The microscopic magnetoelastic tensor for the single ion is given by

$$B_{ijkl}(\text{single ion})=-\beta H_{\mathrm{eff}}S\left(\frac{\partial g_{kl}}{\partial\epsilon_{ij}}\right)_0+CS^2\left(\frac{\partial D_{kl}}{\partial\epsilon_{ij}}\right)_0$$
$$=-\beta H_{\mathrm{eff}}SF_{ijkl}+CS^2G_{ijkl}, \qquad (9)$$

where S is the effective spin of the low-lying manifold and C is a constant of the order of unity given by

$$C=\frac{\langle g|S_z^2-\frac{1}{2}(S_x^2+S_y^2)|g\rangle}{\langle g|S_z^2|g\rangle}, \qquad (10)$$

where $|g\rangle$ represents the ground state. The macroscopic magnetoelastic tensor is related to the microscopic magnetoelastic tensor by

$$B^0{}_{ijkl}=-N\beta H_{\mathrm{eff}}SF_{ijkl}+NCS^2G_{ijkl}, \qquad (11)$$

where N is the number of magnetic ions per unit volume. (This definition is not quite the same as that of Ref. 1, in which the constant C was absorbed into the constant G_{ijkl}.) The parameters $(\partial g_{kl}/\partial\epsilon_{ij})_0=F_{ijkl}$ and $(\partial D_{kl}/\partial\epsilon_{ij})_0=G_{ijkl}$ may be determined for the ions of interest from the strain dependence of the electron-paramagnetic-resonance spectra of the relevant ion in diamagnetic isomorphs of the magnetic crystal of interest. In the present case the appropriate data are that of Ni^{2+}, Mn^{2+}, Fe^{2+}, and Co^{2+}, dilutely incorporated in MgO. Most of the required data are already in the literature of EPR under pressure.

Since we obtain our single-ion strain parameters from a diamagnetic isomorph we lose any explicit information on the strain dependence of H_{eff}. Our presumption is that the exchange field tensor G, which relates H_{eff} to S, has substantially the same topology and strain dependence as the EPR g tensor, and that its explicit omission will introduce errors of perhaps a factor of 2 in magnitude in our magnetoelastic constants but not in their sign or general topology.

We now proceed to utilize the EPR data on Ni^{2+}, Mn^{2+}, Fe^{2+}, and Co^{2+} in MgO to interpret the magnetostriction of NiO, MnO, FeO, and CoO.

IV. NICKEL OXIDE (NiO)

The Ni^{2+} ion has a $3d^8$ configuration which leads to an orbital singlet ground state for octahedral coordination. Spin-orbit coupling does not relieve the threefold spin degeneracy so the lowest-lying state is a spin triplet ($S=1$) with a g factor of 2.227.[15] The crystal-field tensor D is identically zero for the cubic site but has nonvanishing derivatives for deformations from the cubic symmetry. The tensor D is traceless and so there are only two independent single ion constants G_{11} and G_{44} which have been measured by Watkins and Feher[16] to be

$$G_{11}=57\ \mathrm{cm}^{-1}/(\text{unit strain}),$$
$$G_{44}=36\ \mathrm{cm}^{-1}/(\text{unit strain}).$$

We might expect the strain dependence of g also to be significant for Ni^{2+}, but inspection of the data of Germanier and Lacroix[17] leads us to the conclusion that the g shifts under pressure are negligible. We therefore attribute the magnetoelastic constants $\beta^0{}_{ijkl}$ entirely to crystal-field effects

$$B^0{}_{11}=NS^2CG_{11},\quad B^0{}_{44}=NS^2CG_{44}, \qquad (12)$$

where N is the number of Ni^{2+} ions per unit volume of NiO and $S=1$. Using the values of G_{11} and G_{44} cited above and the known elastic constants of MgO[18] we, obtain, for NiO,

$$\lambda_{100}=-1.4\times10^{-4},$$
$$\lambda_{111}=-7.9\times10^{-5}.$$

The spin direction in the (111) plane is believed to be a [211] direction.[19,20] From the above magnetostriction constants we predict a distortion along the spin direction of magnitude

$$\frac{\delta l}{l}[211]=-9.4\times10^{-5}.$$

Since we have assumed all the ions in their lowest energy state, the predicted contraction is implicitly that at 0°K; at higher temperatures, the magnetostriction can

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be found from that at 0°K by using the theory of Callen[21] within the limits stated in that theory.

The in-plane distortion predicted above has been observed by x-ray diffraction techniques. Various authors have reported room-temperature magnitudes for the distortion of 2×10^{-5},[22,23] 9×10^{-5},[20] and 10×10^{-5}.[24] Alberts and Lee[22] confirm that the distortion is a contraction. If the above measurements are extrapolated to 0°K by the theory of Callen,[21] they yield values ranging from 0.4×10^{-4} to 1.8×10^{-4}. The prediction of our model lies close to the average of these values and is of the proper sign.

Yamada[19] has recently proposed that the origin of the magnetostriction in NiO lies primarily in anisotropic exchange and pseudodipolar effects, and that crystal-field effects play only a minor role. Our results are in contradiction to this proposal.

V. MANGANESE OXIDE (MnO)

The Mn^{2+} ion has the $3d^5$ configuration and occupies a spectroscopic ^6S state. Its g value is 2.0014 and essentially strain-independent.[25] The single-ion magnetoelastic constants have been measured by Feher[25] to be

$$G_{11}=1.5\ \text{cm}^{-1}/(\text{unit strain}),$$
$$G_{44}=-0.32\ \text{cm}^{-1}/(\text{unit strain}).$$

By arguments exactly parallel to those used for NiO, we arrive at

$$\lambda_{100}=-3.7\times10^{-5},\quad \lambda_{111}=6.9\times10^{-6}.$$

For an antiferromagnetic spin configuration with the spins lying in the (111) plane, we would have a distortion (in addition to the trigonal exchange striction) at 0°K of

$$(\delta l/l)[211]=-4\times10^{-6}.$$

To our knowledge this distortion has not been observed, and it is probably in fact below the resolution of an x-ray experiment.

VI. FERROUS OXIDE (FeO)

The Fe^{2+} ion has a $3d^6$ configuration which places an orbital triplet lowest in the octahedral field. The fifteen-fold degeneracy of this level (3 orbit×5 spin) is lifted by spin-orbit coupling, leaving a triplet lowest. The g value of this triplet is 3.428[15] and we expect a substantial strain dependence of both g and D in the spin Hamiltonian. Only the strain dependence of D has been reported in the literature. Watkins and Feher's values[16] are

$$G_{11}=800\ \text{cm}^{-1}/(\text{unit strain}),$$
$$G_{44}=540\ \text{cm}^{-1}/(\text{unit strain}).$$

Recently, however, Tucker[26] has made precise measurements of the shift under pressure of the double quantum line of Fe^{2+} in MgO. For this transition, crystal-field shifts cancel out in first order, whereas g shifts do not. Unfortunately, the crystal-field shifts are so large that their contribution in second order is significant, and the raw data must be corrected for these second-order effects before the strain dependence of g may be obtained. Tucker's raw data show that a pressure of 1.38×10^8 dyn/cm² applied along a [110] direction produces shifts of

$$\delta H=-1.059\ \text{Oe when } H_0 \text{ is along } [110],$$
$$\delta H=-3.23\ \text{Oe when } H_0 \text{ is along } [001],$$

where the undisplaced resonance field is 1985 Oe. Using Watkins and Feher's values of G_{11} and G_{44}, we find the correction to $\delta J[110]$ to be +0.66 Oe and the correction to $\delta H[001]$ to be +6.12 Oe, giving

$$\delta H\ (g\ \text{shift})=-0.39\ \text{Oe}\ (H_0\ \text{along}\ (110]),$$
$$\delta H\ (g\ \text{shift})=+2.89\ \text{Oe}\ (H_0\ \text{along}\ [001]).$$

From these field shifts, and assuming $F_{11}=-2F_{12}$ (see below on Co^{2+} in MgO), we obtain

$$F_{11}=-145/(\text{unit strain}),$$
$$F_{44}=39.7/(\text{unit strain}).$$

Finally, using Eqs. (7) and (11), we deduce

$$\lambda_{100}=-4.05\times10^{-2}\ (g\ \text{shift})$$
$$-1.98\times10^{-3}\ (\text{crystal-field effects}),$$
$$\lambda_{111}=+1.0\times10^{-2}\ (g\ \text{shift})$$
$$-1.16\times10^{-3}\ (\text{crystal-field effects}).$$

Note that the contribution to the magnetostriction constants of the g-shift mechanism is in both cases an order of magnitude larger than the crystal-field effects.

The spin direction in FeO is believed to be along the [111] direction perpendicular to the ferromagnetic sheets.[8] We therefore expect a total distortion along this direction of

$$(\delta l/l)[111]=+9.88\times10^{-3}.$$

The value measured in Rooksby and Toombs[4] is $+7.05\times10^{-3}$ at 95°K, in good agreement with the above result. (Extrapolating the measured expansion to 0°K using the theory by Callen[21] gives $\delta l/l=+1.1\times10^{-2}$.) It should be noted that expansion observed overrides the exchange striction which generally produces contractions along the [111] direction at least an order of magnitude smaller than the $\delta l/l$ of FeO.

VII. COBALTOUS OXIDE (CoO)

The Co^{2+} ion has a $3d^7$ configuration which places an orbital triplet lowest in the octahedral field. Spin-orbit coupling splits the twelve-fold degenerate (3 orbit×4

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spin) state leaving a Kramer's doublet lowest. The g value of this doublet in MgO is 4.278.[15]

Since the D term of the spin Hamiltonian vanishes for a doublet state, we expect the magnetoelastic effects to be dominated by the strain dependence of the g tensor. The appropriate measurements have been made by Tucker[27] and are given as

$$F_{11}=-69/(\text{unit strain}),$$
$$F_{12}=32/(\text{unit strain}),$$
$$F_{44}=10/(\text{unit strain}).$$

There are now three independent elements in the F tensor, since the g tensor is not by definition traceless ($F_{11}\approx-2F_{12}$, however). From these parameters we can compute the magnetostriction coefficients using

$$\lambda_{100}=-\frac{2}{3}\frac{\beta H_{\text{eff}}S(F_{11}-F_{12})N}{c_{11}-c_{12}},\quad \lambda_{111}=-\frac{2}{3}\frac{\beta H_{\text{eff}}SF_{44}N}{c_{44}}. \tag{13}$$

We obtain

$$\lambda_{100}=-1.7\times10^{-2},\quad \lambda_{111}=2.2\times10^{-3}$$

using $S=\frac{1}{2}$ and computing H_{eff} from the Néel temperature.

The spin direction in antiferromagnetic CoO is close to the [001][8,28] direction and therefore nearly parallel to the tetragonal distortion (contraction) axis. For this spin orientation we predict substantially the full λ_{100} distortion at 0°K, a large contraction of over 1%. The contraction is large because the small g shift is multiplied by a large effective field, giving a large energy change.

The tetragonal distortion has been measured from the Néel temperature down to 210°K, and must be extrapolated to zero for comparison with our prediction. Slonczewski[29] gives an extrapolated value of -0.8×10^{-2}; we obtain a value of -1.2×10^{-2} using the theory of Callen.[21] Both are in fair agreement with our prediction, having the correct sign and approximately the correct magnitude.

We should not expect exact agreement in this case since we have ignored in our present calculation the effect of a Γ_8 quartet lying only 405 cm^{-1} above the ground Γ_6 doublet. Since we are invoking exchange splittings on the order of 200 cm^{-1}, the interaction of the ground doublet with the excited quartet may be appreciable. We do not believe, however, that the predictions of our model will be substantially altered by this circumstance, since the lowest lying component of the doublet, with which we are primarily concerned, increased its separation from all members of the Γ_8 quartet as the exchange field is increased.

Kanamori[30] has presented a calculation of the magnetostriction of CoO and FeO using a point-ion calculation of the effects of distortions. He achieves apparent agreement with experiment, though the calculation is in several places only semiquantitative.

VIII. CONCLUSIONS

A very simple single-ion molecular-field model predicts correctly the sign and magnitude of the observed magnetostriction in the transition metal monoxides NiO, MnO, FeO, and CoO. The parameters specifying the strain dependence of the crystal-field splittings and the g tensor of the magnetic ions are obtained from the strain dependence of electron-paramagnetic-resonance spectra of the relevant ion in the diamagnetic isomorph MgO.

The model gives a clear physical description of the origin of the magnetostriction. When the wave functions are independent of strain and only the D mechanism is applicable the magnetostriction is small, but whenever the wave functions themselves are sensitive to strain and a g mechanism is present the interaction of the large effective molecular field with the modulated g produces large magnetostrictions.

ACKNOWLEDGMENTS

We are indebted to Dr. Edmund B. Tucker of the General Electric Laboratories for making available to us his data on Fe^{2+} in MgO prior to its publication. We are also indebted to Dr. Ilan Blech of Fairchild Semiconductor Research and Development Laboratories for useful discussions and information on the magnetostriction of NiO.

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