

MAGNETOELASTIC EFFECTS IN EUROPIUM
CHALCOGENIDES

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CHAPTER 1

INTRODUCTION

1.1 Europium Chalcogenides

In recent years a great deal of research has been carried out on the rare earth metals and their compounds. These materials have large spin which gives rise to fascinating magnetic properties. We are particularly interested in the europium chalcogenide series consisting of europium oxide, EuO , europium sulfide EuS , europium selenide EuSe and europium telluride EuTe . These materials are very simple in both crystal and magnetic structure, and are ideal for experimental and theoretical studies. All of them have the NaCl crystal structure as in Fig. 1.1.

The magnetic character of the europium chalcogenide crystals is due to the divalent europium (E^{++}) ion which has the electron configuration $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 5s^2 5p^6 4f^7$. From Hund's rule the seven 4f electrons have their spins parallel, and accordingly the multiplet of lowest energy has $L=0$, $S = \frac{7}{2}$ and $J = \frac{7}{2}$. The spins on the europium atom sites are parallel in EuO , EuS and EuSe ; and these materials are ferromagnetic in character. The direction of magnetization with respect to the crystal axes.

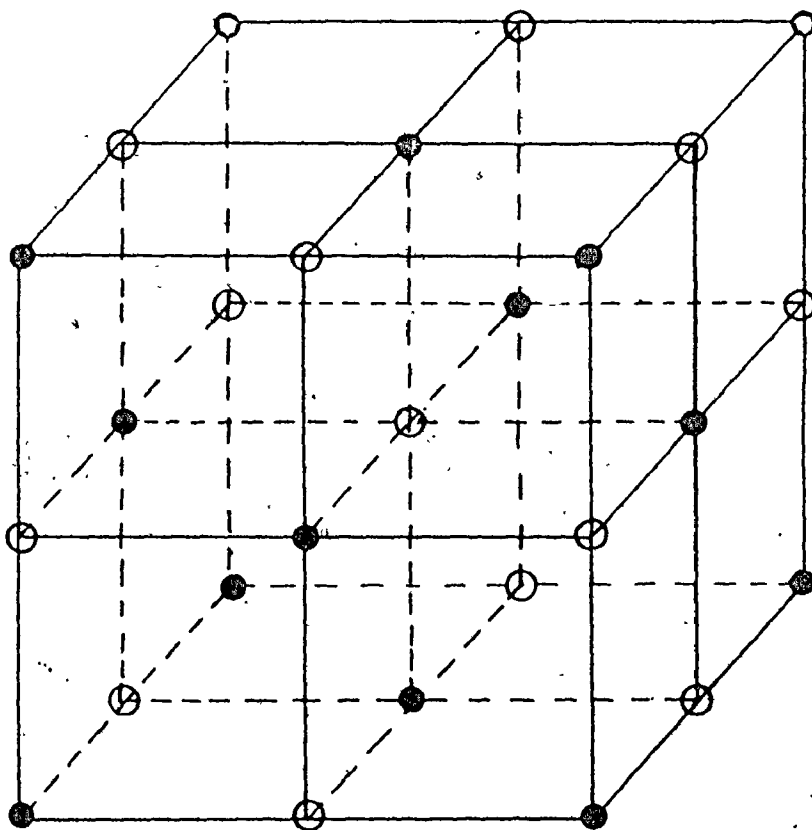


Fig. 1.1 A cubic unit cell of europium chalcogenide.

- are europium ions
- are chalcogenide ions

is determined by the magnetocrystalline anisotropy, which has its origin in the crystalline electric field acting at the Eu sites. Thus there is a preferred direction for the magnetization in each crystal, but it need not be the same for different members of the chalcogenide series. On the other hand EuTe is antiferromagnetic, the spins being parallel in planes perpendicular to the $[111]$ direction and the spin directions alternating from one (111) plane of magnetic ions to the next. The preferred orientation of the spins in a particular (111) plane depends on a relatively weak anisotropy energy in this plane.

1.2 Previous Works on Magnetoelastic Interactions

The phenomenon of 'magnetostriction', which describes the elastic deformation of a magnetic substance under an applied magnetic field, has been known for many years (see, for example, Bozorth, 1951, and references therein). But it was not until 1958 that the phenomenological study of 'magnetoelastic interactions' began with the independent works of Kittel (1958) and Akhiezer et al. (1958). They described the magnetoelastic interactions as a coupling of the elastic strains with the magnetic spins. The 'finite strain theory' for a magnetoelastic system was later developed through the works of Toupin (1956), Tiersten

(1964) and Brown (1964,1965,1966). In contrast to the small strain theory of Kittel et al. in this theory the total angular momentum (spin plus lattice) of the combined magnetic and elastic systems is conserved, i.e., the Hamiltonian of the combined system is rotationally invariant. Eastman (1966) experimentally measured the magnetoelastic effects in yttrium iron garnet (YIG), and he was able to interpret the results using the finite strain theory. The measurement of the change in elastic constants of MnF_2 as a function of an applied magnetic field by Melcher (1970) also demonstrated the validity of the finite strain theory for an antiferromagnet. The rotationally invariant theory of magnetoelastic interactions in the heavy rare earth metals was developed by Southern and Goodings (1973) and they predicted the fractional change in elastic constants for Gd, Tb, Dy, Ho, and Er in both the ferromagnetic and antiferromagnetic regions. The subsequent measurements, of the dependence of certain elastic constants as a function of applied magnetic field for single crystal Tb in the paramagnetic region, by Salama et al. (1973) were in reasonably good agreement with the predictions of Southern and Goodings. The theory for ferromagnetic crystals of cubic symmetry has been formulated by Southern (1973,1973a) and he has predicted the change in elastic constants as a function of applied magnetic field for several rare earth iron cubic Laves phase compounds.

1.3 Scope of the Thesis

In the present work we describe the theory of magnetoelastic interactions for a ferromagnetic cubic crystal and apply it to the europium chalcogenides to calculate the change in elastic constants as a function of the applied magnetic field. In Chapter 2 we begin by discussing a simple Hamiltonian for a magnetic system, which consists of the Heisenberg exchange term and the interaction of the spins with an applied magnetic field. The spin waves, which are the low-lying excited states of the magnetic system, are briefly discussed. The crystal field terms for cubic symmetry are then introduced and the magnetocrystalline anisotropy is described, the anisotropy constants being expressed in terms of the crystal field parameters.

In Chapter 3 we describe the elastic system. The 'finite strain theory' and 'small strain theory' for the elastic deformation of a medium are discussed briefly. Then we write down the elastic Hamiltonian of a crystal having cubic symmetry.

In Chapter 4 we consider the coupling between the magnetic and elastic systems. The magnetoelastic coupling Hamiltonian for single ion interactions in a cubic ferromagnetic crystal can be written in terms of magnetoelastic constants, symmetry strains and tensor

operators of the spins. The total Hamiltonian of a magnetoelastic system is the combination of magnetic, elastic and magnetoelastic coupling Hamiltonians. We discuss also the magnetostriction, the relation between the magnetostriction constants and the magnetoelastic constants, and the magnetoelastic modes that result from the presence of the magnetoelastic coupling. From these coupled modes one can obtain an expression for the effective elastic constant of a cubic crystal as a function of the applied magnetic field.

In Chapter 5 we apply the theory to the europium chalcogenide series. We tabulate their basic magnetic data, and experimental values of anisotropy constants and magnetostriction coefficients. From these values the crystal field parameters and the magnetoelastic constants for EuO are estimated. Finally we compute the change in various elastic constants of EuO as a function of magnetic field, when the magnetic field is applied along different symmetry directions. The results are then compared with similar calculations for the rare earth iron cubic Laves compounds.

CHAPTER 2
THE MAGNETIC SYSTEM

All magnetic substances have magnetic moments associated with each individual atom. The magnetic moment of a free atom comes from the total spin of its electrons and their total orbital angular momentum about the nucleus. Since the magnetic moment of the nucleus is approximately 10^{-3} times smaller than the magnetic moment due to the electrons, then it can usually be neglected. The magnetic moment associated with total orbital angular momentum \vec{L} is (in the MKS system):

$$[2.1] \quad \vec{\mu}_L = - \frac{e\hbar}{2m_e} \frac{\vec{L}}{\hbar}$$

and that associated with total spin angular momentum \vec{S} is

$$[2.2] \quad \vec{\mu}_S = - 2 \frac{e\hbar}{2m_e} \frac{\vec{S}}{\hbar} .$$

The quantity $\mu_B = \frac{e\hbar}{2m_e}$ is called the Bohr magneton. In terms of the total angular momentum \vec{J} the magnetic moment is given by

$$[2.3] \quad \vec{\mu} = - g\mu_B \vec{J} ,$$

where

$$[2.4] \quad g = 1 + \frac{J(J+1)S(S+1) - L(L+1)}{2J(J+1)}$$

is the Lande g-factor.

The magnetization \vec{M} is defined as the magnetic moment per unit volume. When an external magnetic field is applied on a magnetic substance magnetic moments corresponding to individual atoms tend to align themselves in the direction of the applied field, and a resultant magnetization occurs. As the external field increases the magnetization increases upto a saturation point due to the alignment of domains. In the case of ferromagnets a spontaneous magnetization can exist in the absence of any external magnetic field.

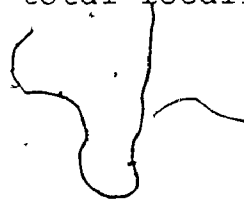
Within a single domain the magnetization decreases as the temperature increases, and eventually goes to zero at the Curie temperature T_c .

In compounds of the rare earth elements the interactions between magnetic moments are well described by the Heisenberg Hamiltonian:

$$[2.5] \quad \mathcal{H} = - \sum_{i < j} J_{ij} \vec{S}_i \cdot \vec{S}_j$$



Here \vec{S}_i represents the total angular momentum of the i^{th} atom, and the exchange integral J_{ij} depends on the relative positions of the atoms. Specially for our works in europium compounds, since europium atom has zero orbital angular momentum, \vec{S}_i represents the total localized spin of the i^{th} atom.



2.1 Spin Waves

In the ground state, for ferromagnets, all spins are parallel, and have the maximum allowable value for the spin component in the direction of magnetization. The low-lying excited states are given by the linear combination of ferromagnetic states, each with a single spin flipped and modulating these by a phase factor. Such an excitation is called a spin wave or, when quantized, a magnon.

When viewed in a semiclassical way, which allows simple pictorial interpretations, as in Fig. 2.1, the low-lying elementary excitations consist of the spins precessing about the direction of magnetization. Due to interactions the successive spins advance in phase by a constant angle. The wavelike form of elementary excitations is referred to as a spin wave.

In the presence of an applied magnetic field, \vec{H} , the energy of interaction with this field is $-\vec{\mu} \cdot \vec{H}$, where

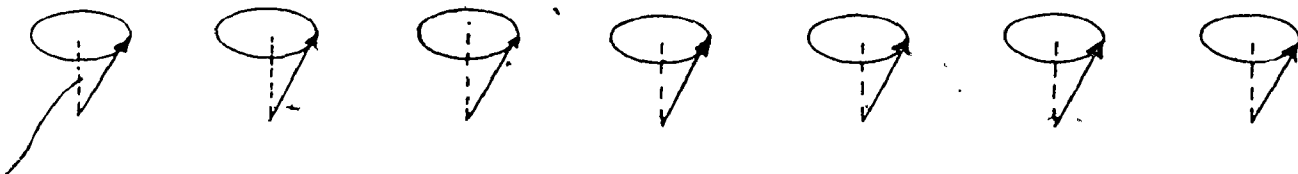


Fig. 2.1(a) Ground state of ferromagnets.



Fig. 2.1(b) Precession of the spins in phase.



Fig. 2.1(c) Wavelike form of the precession of spins when viewed from above.

the magnetic moment is given by

$$[2.6] \quad \vec{\mu} = -g\mu_B \sum_i \vec{S}_i .$$

Therefore, the Hamiltonian of the magnetic system, in the presence of the field \vec{H} , is

$$[2.7] \quad \mathcal{H} = - \sum_{i < j} J_{ij} \vec{S}_i \cdot \vec{S}_j + g\mu_B \vec{H} \cdot \sum_i \vec{S}_i .$$

When a spin wave is created, involving the reversal of one unit of spin, an additional energy $g\mu_B H$ must be supplied, and hence this introduces an energy gap in the spin wave spectrum.

In addition to an applied magnetic field there may also exist an effective 'anisotropy field', \vec{H}_A , which may arise from crystalline electric fields or from dipole-dipole interactions. Although not strictly speaking a magnetic field, the anisotropy energy can often be represented by an effective field \vec{H}_A which also introduces a gap in the spin wave spectrum. We will discuss the 'anisotropy energy' in the next section.

The standard theory of spin waves (see, for example, Kittel, 1971) yields the following result for the energy of a spin wave of wavevector \vec{q} (for a ferromagnetic cubic lattice with nearest neighbor interactions):

$$[2.8] \quad E_{\vec{q}} = E_0 + g\mu_B \vec{H} + 2JS \left[Z - \sum_{\delta} \cos(\vec{q} \cdot \vec{\delta}) \right]$$

Here E_0 is the ground state energy,

J is the nearest neighbor exchange integral J_{ij} ,

S is the magnitude of spin component in the direction of magnetization,

Z is the number of nearest neighbors, and

$\vec{\delta}$ is a nearest neighbor vector.

At small q (long wavelength) the excitation energy is ($qa \ll 1$, where a is the lattice constant):

$$\approx g\mu_B H + 2JSa^2 q^2$$

As q goes to zero excitation energy go to a constant value $g\mu_B H$. Due to the presence of any anisotropy energy there may be an additional constant term with $g\mu_B H$. The excited low frequency spin waves give the spin wave resonance. With certain applied fields $q=0$ spin waves are excited and we get the spin wave resonance.

2.2 Magnetocrystalline Anisotropy Energy

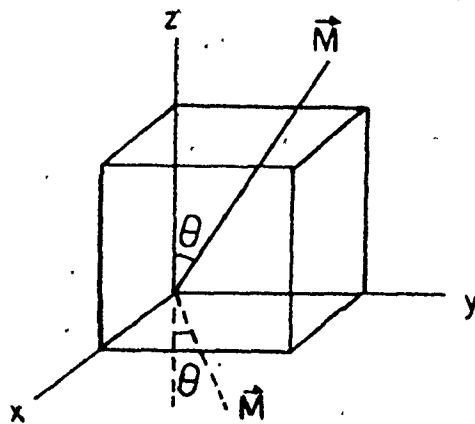
Experimental measurements reveal that ferromagnetic or ferrimagnetic single crystals can be magnetized most easily by an applied magnetic field in certain directions

called easy directions of magnetization. Hence it follows that there is an energy in the ferromagnetic crystal which favours the magnetization lying along the easy directions. This energy is called the magnetocrystalline anisotropy energy. This anisotropy energy depends only on the direction of magnetization. For a saturated crystal the anisotropy energy density may be formally written as

$$[2.9] \quad F = b_i \alpha_i + b_{ij} \alpha_i \alpha_j + b_{ijk} \alpha_i \alpha_j \alpha_k + \dots$$

Here repeated indices denote summation, the α_i are the direction cosines of the magnetization with respect to the crystal axes; b_i , b_{ij} , etc. are constant coefficients. One can expand F in ascending powers of α_i in the hope that higher order terms are small.

For a cubic crystal if the magnetization makes



an angle θ with the z-axis, $\alpha_3 = \cos\theta$. Also if the magnetization makes an angle $\pi - \theta$ with the z-axis, then because of the cubic symmetry the free energy must remain

the same. However, because

$$[2.10] \quad \alpha_3 = \cos(\pi - \theta) = -\cos\theta,$$

therefore, all odd terms in α_3 must vanish. Since in a cubic crystal all the three axes x, y, z are equivalent, therefore, odd terms in α_1 and α_2 must also vanish. Also we know that

$$[2.11] \quad \alpha_1^2 + \alpha_2^2 + \alpha_3^2 = 1.$$

Now expanding F in terms of α_i , taking into consideration the above symmetry requirements and algebraic relationships we come to the form

$$[2.12] \quad F = K_0 + K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) + K_2\alpha_1^2\alpha_2^2\alpha_3^2,$$

where K_1 and K_2 are called the first and second anisotropy constants. Putting

$$[2.13a] \quad \alpha_1 = \sin\theta\cos\phi$$

$$[2.13b] \quad \alpha_2 = \sin\theta\sin\phi$$

$$[2.13c] \quad \alpha_3 = \cos\theta$$

we get

$$\begin{aligned}
 [2.14] \quad F = & K_0 + K_1 \sin^2 \theta - \frac{1}{8} (7K_1 - K_2) \sin^4 \theta \\
 & - \frac{1}{8} (K_1 + K_2) \sin^4 \theta \cos 4\phi - \frac{1}{8} K_2 \sin^6 \theta \\
 & + \frac{1}{8} K_2 \sin^6 \theta \cos 4\phi .
 \end{aligned}$$

For hexagonal crystals the anisotropy energy is found by a similar analysis to have the form

$$\begin{aligned}
 [2.15] \quad F = & K_0 + K_1 \sin^2 \theta + K_2 \sin^4 \theta + K_3 \sin^6 \theta \\
 & + K_4 \sin^6 \theta \cos 6\phi ,
 \end{aligned}$$

where θ is the angle between the magnetization and the hexagonal x_3 -axis.

It is more convenient to express the anisotropy energy in terms of the (unnormalized) spherical harmonics, $\tilde{Y}_{\ell m}$;

$$[2.16] \quad F = \sum_{\ell, m} k_{\ell m} \tilde{Y}_{\ell m}$$

where $k_{\ell m}$ are called anisotropy coefficients. We will express them in terms of anisotropy constants. We take the spherical harmonics as

$$[2.17] \quad \tilde{Y}_{\ell m} = \left[\frac{(\ell-m)!}{(\ell+m)!} \right]^{\frac{1}{2}} p_{\ell}^{|m|} (\cos\theta) e^{im\phi},$$

where the factor $(-1)^m$ is included for $m > 0$,

and not included for $m < 0$.

In terms of these $\tilde{Y}_{\ell m}$ the anisotropy energy for cubic crystals has the form

$$[2.18] \quad F = K_0 + \frac{1}{105} (21K_1 + K_2) \tilde{Y}_{00} - \frac{1}{55} (11K_1 + K_2) \tilde{Y}_{40} \\ - \frac{1}{11\sqrt{70}} (11K_1 + K_2) [\tilde{Y}_{44} + \frac{1}{8!} \tilde{Y}_{4,-4}] \\ + \frac{2}{231} K_2 \tilde{Y}_{60} - \frac{2\sqrt{7}}{231} K_2 [\tilde{Y}_{64} + \frac{1}{10!} \tilde{Y}_{6,-4}]$$

For hexagonal crystals the anisotropy energy has the form

$$[2.19] \quad F = K_0 + \frac{2}{105} (35K_1 + 28K_2 + 24K_3) \tilde{Y}_{00} \\ - \frac{2}{21} (7K_1 + 8K_2 + 8K_3) \tilde{Y}_{20} \\ + \frac{8}{385} (11K_2 + 18K_3) \tilde{Y}_{40} - \frac{16}{231} K_3 \tilde{Y}_{60} \\ + \frac{16}{3} \sqrt{\frac{2}{77}} K_4 [\tilde{Y}_{66} + \frac{1}{12!} \tilde{Y}_{6,-6}]$$

2.3 The Tensor Operators $O_{\ell m}$

Up to this point the expression for magnetocrystalline anisotropy is purely classical, depending on the direction of magnetization through the angles θ and ϕ occurring in $Y_{\ell m}$. While it is possible to obtain the main results of this thesis using classical mechanics, following Goodings and Southern (1971) we have chosen to use the techniques of quantum mechanics. This requires expressing the anisotropy energy in terms of the spin operators S_i at each site.

To obtain operator equivalents we follow Steven's prescription (Stevens, 1952; Hutchings, 1964). We first write $Y_{\ell m}$ in terms of cartesian coordinates x, y, z ; then x, y, z are replaced by S_x, S_y, S_z respectively always allowing for the noncommutation for S_x, S_y, S_z . This is done by replacing products of x, y, z by an expression consisting of all the possible different orderings of the corresponding operators S_x, S_y, S_z and divided by the total number of such terms. The newly formed operator is written in terms of the operators $\tilde{O}_{\ell m}$ tabulated by Buckmaster (1962). These operators $\tilde{O}_{\ell m}$ have the same transformation properties under rotations as those of $\tilde{Y}_{\ell m}$.

In terms of the $\tilde{O}_{\ell m}$ the anisotropy energy for a cubic crystal has the form (Southern, 1973),

$$\begin{aligned}
 [2.20] \quad F = & \tilde{B}_4^0 \sum_i [\tilde{O}_{40}(S_i) + (\frac{10}{7})^{\frac{1}{2}} \tilde{O}_{44}^+(S_i)] \\
 & + \tilde{B}_6^0 \sum_i [\tilde{O}_{60}(S_i) - (14)^{\frac{1}{2}} \tilde{O}_{64}^+(S_i)]
 \end{aligned}$$

The $\tilde{O}_{\ell m}^+$ are related to the operators $O_{\ell m}$ as follows:

$$[2.21a] \quad \tilde{O}_{\ell m}^+ = \frac{1}{2} (O_{\ell m} + O_{\ell -m})$$

$$[2.21b] \quad \tilde{O}_{\ell m}^- = \frac{1}{-2i} (O_{\ell m} - O_{\ell -m})$$

\tilde{B}_4^0 and \tilde{B}_6^0 are crystal field parameters which are related to the anisotropy constants K_1 and K_2 introduced in the last section. Now combining the exchange term, the Zeeman term and the magnetocrystalline anisotropy energy we can write the Hamiltonian of a magnetic system for a cubic crystal as:

$$\begin{aligned}
 [2.22] \quad \mathcal{H}_m = & - \sum_{i < j} J_{ij} \vec{S}_i \cdot \vec{S}_j + g\mu_B \vec{H} \cdot \sum_i \vec{S}_i \\
 & + \tilde{B}_4^0 \sum_i [\tilde{O}_{40}(S_i) + (\frac{10}{7})^{\frac{1}{2}} \tilde{O}_{44}^+(S_i)] \\
 & + \tilde{B}_6^0 \sum_i [\tilde{O}_{60}(S_i) - (14)^{\frac{1}{2}} \tilde{O}_{64}^+(S_i)].
 \end{aligned}$$

In a similar way we get the Hamiltonian of a magnetic system for a hexagonal crystal as (Goodings and Southern, 1971),

$$\begin{aligned}
 [2.23] \quad \mathcal{L}_m = & - \sum_{i < j} J_{ij} \vec{S}_i \cdot \vec{S}_j + g \mu_B \vec{H} \cdot \sum_i \vec{S}_i \\
 & + \tilde{B}_2^0 \sum_i \tilde{O}_{20}(S_i) + \tilde{B}_4^0 \sum_i \tilde{O}_{40}(S_i) \\
 & + \tilde{B}_6^0 \sum_i \tilde{O}_{60}(S_i) + \tilde{B}_6^6 \sum_i [\tilde{O}_{66}(S_i) + \tilde{O}_{6-6}(S_i)] .
 \end{aligned}$$

CHAPTER 3
THE ELASTIC SYSTEM

We discuss the elastic deformation of a medium in this chapter. The basic idea is that any elastic deformation can be analyzed into a 'stretching' or an 'elongation' plus a rotation of the medium. In the finite strain theory the stretching is described by the finite strain tensor E_{ij} and the rotation is described by the finite rotation tensor R_{ij} .

3.1 Finite Strain Tensor $E_{\mu\nu}$

We consider an elastic medium which experiences a deformation. In the initial or undeformed state the coordinates of a mass element with respect to any convenient rectangular cartesian reference frame are denoted by X_μ ($\mu = 1, 2, 3$) as in Fig. 3.1. Similarly in the final or deformed state, the coordinates of the same mass element with respect to any other convenient reference frame are denoted by x_μ ($\mu = 1, 2, 3$). Also dX_μ goes to dx_μ . The deformation of a point is described by the relation of the coordinates of the same mass element in the

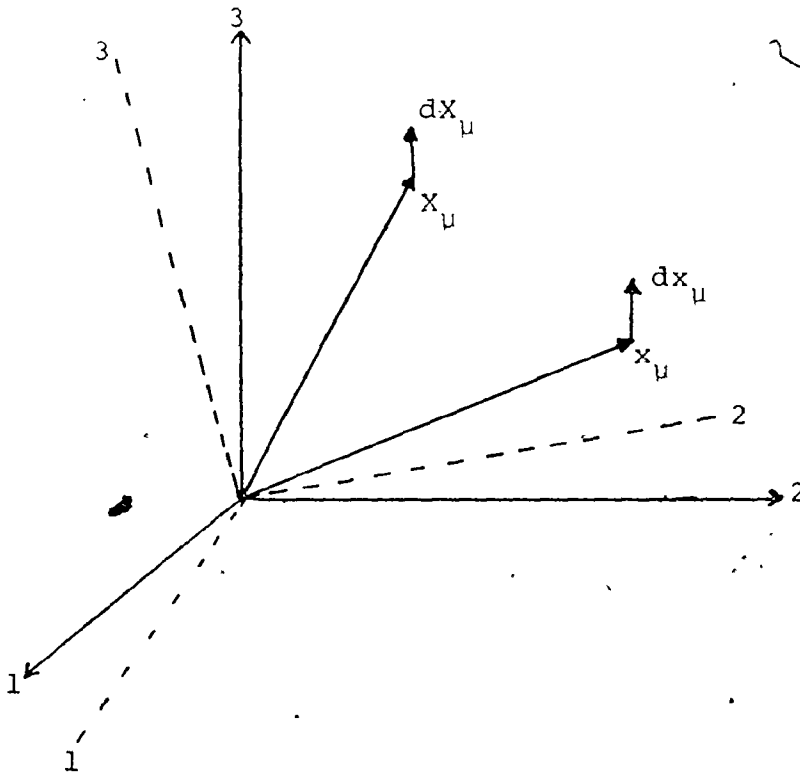


Fig. 3.1 Deformation of a mass element.

undeformed and deformed states. The relation may be written as

$$[3.1a] \quad x_{\mu} = x_{\mu}(X_{\nu})$$

or

$$[3.1b] \quad X_{\mu} = X_{\mu}(x_{\nu})$$

The X_{μ} are called the material or Lagrangian coordinates and a material description uses these as the independent variables. A spatial description uses the x_{μ} as independent variables and these are referred to as spatial or Eulerian coordinates. The deformation may be described using either method, but we will use the material description since the undeformed state has certain known symmetry properties.

After deformation the change in square of a length in the medium is

$$dx_{\mu} dx_{\mu} - dX_{\mu} dX_{\mu}$$

Since $x_{\mu} = x_{\mu}(X_{\nu})$,

$$[3.2] \quad dx_{\mu} = \frac{\partial x_{\mu}}{\partial X_{\nu}} dX_{\nu}$$

Summation over ν is implied here and $\partial x_\mu / \partial X_\nu$ is called the deformation gradient. Now the change in the square of the length is

$$\begin{aligned}
 [3.3] \quad dx_\lambda dx_\lambda - dX_\lambda dX_\lambda &= \frac{\partial x_\lambda}{\partial X_\mu} \frac{\partial x_\lambda}{\partial X_\nu} dX_\mu dX_\nu - \delta_{\mu\nu} dX_\mu dX_\nu \\
 &= \left(\frac{\partial x_\lambda}{\partial X_\mu} \frac{\partial x_\lambda}{\partial X_\nu} - \delta_{\mu\nu} \right) dX_\mu dX_\nu \\
 &= 2E_{\mu\nu} dX_\mu dX_\nu
 \end{aligned}$$

This defines the finite strain tensor as

$$[3.4] \quad E_{\mu\nu} = \frac{1}{2} \left(\frac{\partial x_\lambda}{\partial X_\mu} \frac{\partial x_\lambda}{\partial X_\nu} - \delta_{\mu\nu} \right)$$

It is easily seen that $E_{\mu\nu}$ is a symmetric tensor which, because of its scalar-product form, does not depend on the choice of reference frame employed for the deformed state.

We introduce the displacement

$$[3.5] \quad u_\lambda = x_\lambda - X_\lambda$$

Therefore,

$$[3.6a] \quad \frac{\partial u_\lambda}{\partial X_\mu} = \frac{\partial x_\lambda}{\partial X_\mu} - \delta_{\mu\lambda}$$

or

$$[3.6b] \quad \frac{\partial x_\lambda}{\partial X_\mu} = \frac{\partial u_\lambda}{\partial X_\mu} + \delta_{\mu\lambda}$$

Putting this in $E_{\mu\nu}$ we get

$$[3.7] \quad E_{\mu\nu} = \frac{1}{2} \left(\frac{\partial u_\nu}{\partial X_\mu} + \frac{\partial u_\mu}{\partial X_\nu} \right) + \frac{1}{2} \frac{\partial u_\lambda}{\partial X_\mu} \frac{\partial u_\lambda}{\partial X_\nu}$$

$$\equiv \varepsilon_{\mu\nu} + \frac{1}{2} \frac{\partial u_\lambda}{\partial X_\mu} \frac{\partial u_\lambda}{\partial X_\nu}$$

$\varepsilon_{\mu\nu}$ is the symmetric strain function of the usual small strain theory. The second term on the right hand side is of second order in the displacement gradients and is neglected in the small strain theory.

3.2 Finite Rotation Tensor $R_{\mu\nu}$

We define a tensor $C_{\mu\nu}$ - called Green's deformation tensor as:

$$[3.8] \quad C_{\mu\nu} = \frac{\partial x_\lambda}{\partial X_\mu} \frac{\partial x_\lambda}{\partial X_\nu}$$

From the definition of the finite strain tensor we find,

$$[3.9] \quad C_{\mu\nu} = 2E_{\mu\nu} + \delta_{\mu\nu}$$

$C_{\mu\nu}$ is a measure of the strain. This is seen from the expressions for the square of the distance between two neighboring points before and after the deformation. The square of the distance before the deformation is

$$[3.10] \quad ds_0^2 = dx_\lambda dx_\lambda$$

and that after the deformation is

$$[3.11] \quad ds^2 = dx_\lambda dx_\lambda = \frac{\partial x_\lambda}{\partial X_\mu} \frac{\partial x_\lambda}{\partial X_\nu} dX_\mu dX_\nu$$

$$= C_{\mu\nu} dX_\mu dX_\nu$$

When $E_{\mu\nu} = 0$, from Eq. [3.9], $C_{\mu\nu} = \delta_{\mu\nu}$ and $ds^2 = ds_0^2$, i.e., the neighborhood under consideration has undergone at most a rigid body displacement and a rotation. Also from Eq. [3.11], we see that $C_{\mu\nu} dX_\mu dX_\nu$ is positive definite. We also see from the definition that $C_{\mu\nu}$ is symmetric; it therefore represents only 6 independent quantities instead of 9 quantities represented by deformation gradients.

The symmetric matrix $C_{\mu\nu}$ can be brought to diagonal form by an orthogonal transformation. We can write

$$[3.12] \quad S_{\lambda\mu} C_{\mu\nu} S_{\nu\lambda}^{-1} = C_\lambda \delta_{\lambda\lambda}$$

where $S_{\lambda\mu}$ is an orthogonal matrix. Let the diagonal

elements in this representation be C_1, C_2, C_3 ; they are all positive. Then the symmetric tensor with the same principal axes but with elements $C_1^{\frac{1}{2}}, C_2^{\frac{1}{2}}, C_3^{\frac{1}{2}}$ may be defined as the tensor $(C^{\frac{1}{2}})_{\mu\nu}$, i.e., following

$$[3.13] \quad C_{\mu\nu} = S_{\mu\lambda}^{-1} C_{\lambda} S_{\lambda\nu}$$

we can write

$$[3.14] \quad (C^{\frac{1}{2}})_{\mu\nu} = S_{\mu\lambda}^{-1} C_{\lambda}^{\frac{1}{2}} S_{\lambda\nu}$$

Then we see that

$$\begin{aligned}
 [3.15] \quad (C^{\frac{1}{2}})_{\mu\zeta} (C^{\frac{1}{2}})_{\zeta\nu} &= S_{\mu\lambda}^{-1} C_{\lambda}^{\frac{1}{2}} S_{\lambda\zeta} S_{\zeta\lambda}^{-1} C_{\lambda}^{\frac{1}{2}} S_{\lambda\nu} \\
 &= S_{\mu\lambda}^{-1} C_{\lambda}^{\frac{1}{2}} \delta_{\lambda\lambda} C_{\lambda}^{\frac{1}{2}} S_{\lambda\nu} \\
 &= S_{\mu\lambda}^{-1} C_{\lambda} S_{\lambda\nu}
 \end{aligned}$$

or

$$[3.16] \quad (C^{\frac{1}{2}})_{\mu\zeta} (C^{\frac{1}{2}})_{\zeta\nu} = C_{\mu\nu}$$

We now define a tensor

$$[3.17] \quad R_{\mu\lambda} \equiv x_{\lambda,\nu} (C^{\frac{1}{2}})_{\mu\nu}$$

where $C^{-\frac{1}{2}}$ is defined in the principal axes in a manner similar to the definition of $C^{\frac{1}{2}}$. Rotated back to the original coordinate system it is $(C^{-\frac{1}{2}})_{\mu\nu}$. We can easily see using the definition of $C_{\mu\nu}$ in Eq. [3.8] that

$$[3.18] \quad R_{\mu\lambda} R_{\gamma\lambda} = \delta_{\mu\gamma}$$

i.e., $R_{\mu\lambda}$ is an orthogonal matrix and describes a rigid body rotation of the neighborhood under consideration.

Inverting the Eq. [3.17], we find,

$$[3.19] \quad x_{\lambda,\mu} = (C^{\frac{1}{2}})_{\mu\nu} R_{\nu\lambda}$$

which analyzes the deformation into a finite rotation followed by a finite strain. The three Euler angles equivalent to R , plus the 6 components of C , are together equivalent to the 9 components of $x_{\lambda,\mu}$.

3.3 $E_{\mu\nu}$ and $R_{\mu\nu}$ in Small Strain Theory

In small strain theory we write the displacement

$$[3.20] \quad u_{\lambda} = x_{\lambda} - X_{\lambda}$$

The displacement gradient is

$$[3.21] \quad u_{\lambda, \mu} = x_{\lambda, \mu} - \delta_{\lambda \mu}$$

The $u_{i, j}$ is generally written as

$$[3.22] \quad u_{i, j} = \frac{1}{2} (u_{i, j} + u_{j, i}) + \frac{1}{2} (u_{i, j} - u_{j, i})$$

$$\equiv \epsilon_{ij} + \omega_{ij}$$

We have already encountered the symmetric strain function ϵ_{ij} ; ω_{ij} is the antisymmetric strain function and represents a rotation of the medium as a whole without any deformation.

Now from Eq. [3.4],

$$[3.23] \quad E_{\mu\nu} = \frac{1}{2} (x_{\lambda, \mu} x_{\lambda, \nu} - \delta_{\mu\nu})$$

$$= \frac{1}{2} [(u_{\lambda, \mu} + \delta_{\lambda\mu})(u_{\lambda, \nu} + \delta_{\lambda\nu}) - \delta_{\mu\nu}]$$

$$= \frac{1}{2} (u_{\mu, \nu} + u_{\nu, \mu}) + \frac{1}{2} u_{\lambda, \mu} u_{\lambda, \nu}$$

$$= \epsilon_{\mu\nu} + \frac{1}{2} (\epsilon_{\lambda\mu} + \omega_{\lambda\mu})(\epsilon_{\lambda\nu} + \omega_{\lambda\nu})$$

$$= \epsilon_{\mu\nu} + \frac{1}{2} (\epsilon_{\lambda\mu} \epsilon_{\lambda\nu} + \epsilon_{\lambda\mu} \omega_{\lambda\nu} + \omega_{\lambda\mu} \epsilon_{\lambda\nu} + \omega_{\lambda\mu} \omega_{\lambda\nu})$$

We also have the Eq. [3.9],

$$C_{\mu\nu} = \delta_{\mu\nu} + 2E_{\mu\nu} .$$

Now if $C^{-\frac{1}{2}}$ is the diagonal matrix in the principal axes, we can write

$$\begin{aligned} [3.24] \quad C^{-\frac{1}{2}} &= (1 + 2E)^{-\frac{1}{2}} \\ &= 1 - E + \frac{3}{2} E^2 + \dots , \end{aligned}$$

where this expansion is carried out separately for each of the diagonal elements of $C^{-\frac{1}{2}}$. Then we make a rotation with matrix R^{-1} :

$$[3.25] \quad R^{-1} C^{-\frac{1}{2}} R = R^{-1} R - R^{-1} E R + \frac{3}{2} (R^{-1} E R) (R^{-1} E R) + \dots$$

or

$$\begin{aligned} [3.26] \quad (C^{-\frac{1}{2}})_{\mu\nu} &= \delta_{\mu\nu} - \frac{1}{2} (u_{\mu,\nu} + u_{\nu,\mu}) - \frac{1}{2} u_{\lambda,\mu} u_{\lambda,\nu} \\ &\quad + \frac{3}{2} \cdot \frac{1}{2} (u_{\mu,\alpha} + u_{\alpha,\mu}) \frac{1}{2} (u_{\alpha,\nu} + u_{\nu,\alpha}) , \end{aligned}$$

up to second order in displacement gradients. Using the definition of $\epsilon_{\mu\nu}$ we can write

$$[3.27] \quad (C^{-\frac{1}{2}})_{\mu\nu} = \delta_{\mu\nu} - \epsilon_{\mu\nu} - \frac{1}{2} u_{\lambda,\mu} u_{\lambda,\nu} + \frac{3}{2} \epsilon_{\mu\alpha} \epsilon_{\alpha\nu} .$$

Working through we find up to the second order in displacement gradients

$$\begin{aligned}
 [3.28] \quad R_{\mu\lambda} &= \delta_{\mu\lambda} + \omega_{\lambda\mu} - u_{\lambda,\nu} \varepsilon_{\mu\nu} - \frac{1}{2} u_{\alpha,\mu} u_{\alpha,\nu} \\
 &\quad + \frac{3}{2} \varepsilon_{\mu\alpha} \varepsilon_{\alpha\lambda} .
 \end{aligned}$$

In the first order approximation this leads to

$$[3.29] \quad R_{\mu\lambda} = \delta_{\mu\lambda} + \omega_{\lambda\mu} .$$

This leads to the result in first order displacement gradients as follows:

$$[3.30] \quad (C^{\frac{1}{2}})_{\mu\nu} = \delta_{\mu\nu} + \varepsilon_{\mu\nu}$$

and

$$\begin{aligned}
 [3.31] \quad x_{\lambda,\mu} &= (C^{\frac{1}{2}})_{\mu\nu} R_{\nu\lambda} \\
 &= (\delta_{\mu\nu} + \varepsilon_{\mu\nu}) (\delta_{\lambda\nu} + \omega_{\lambda\nu}) \\
 &= (\delta_{\nu\mu} + \varepsilon_{\nu\mu}) (\delta_{\lambda\nu} + \omega_{\lambda\nu}) \\
 &= \delta_{\lambda\mu} + \varepsilon_{\lambda\mu} + \omega_{\lambda\mu} .
 \end{aligned}$$

Therefore the displacement gradient

$$[3.32] \quad u_{\lambda, \mu} = x_{\lambda, \mu} - \delta_{\lambda\mu} = \epsilon_{\lambda\mu} + \omega_{\lambda\mu} .$$

But, from the definition of $u_{\lambda, \mu}$, we know that this is an exact expression. Therefore, we may conclude that the two expressions for C^2 and R , when multiplied together, must cancel in higher order terms.

3.4 The Hamiltonian of the Elastic System \mathcal{H}_e

We assume the elastic medium to be constrained so that all 'antisymmetric strains' such as ω_{ij} , which correspond to homogeneous rotations of the system, vanish. The elastic energy \mathcal{H}_e is then obtained in terms of the symmetric strain components E_{xx} , E_{yy} , E_{zz} , E_{xy} , E_{yz} and E_{zx} . Following Callen and Callen (1965) we see that, under the crystal symmetry operations of the point group G , the six strain components transform into each other and thereby generate a six-dimensional representation of G . This representation is reducible. In particular, the quantity $E^\alpha \equiv (E_{xx} + E_{yy} + E_{zz})$ always transforms under the fully symmetric representation Γ_α of G . The remaining five-dimensional representation is further reduced by the linear combinations $[E_{zz} - \frac{1}{3}(E_{xx} + E_{yy} + E_{zz})]$, $(E_{xx} - E_{yy})$, E_{xy} ,

E_{yz}, E_{xz} . In terms of these strain functions the energy associated with the elastic medium is written, in a general form, as

$$[3.33] \quad \mathcal{H}_e = \sum_{\Gamma} \sum_{j,j'} \frac{1}{2} C_{jj}^{\Gamma} \sum_i E_i^{\Gamma,j} E_i^{\Gamma,j'}$$

where Γ labels the irreducible representation of order n and i labels the basis functions ($i = 1, 2, \dots, n$). As there may be more than one such set we distinguish them by the superscripts j . The symmetry elastic constants C_{jj}^{Γ} are linear combinations of the usual cartesian elastic constants.

For the case of cubic symmetry the elastic Hamiltonian \mathcal{H}_e takes the form (see Southern, 1973a, 97)

$$[3.34] \quad \mathcal{H}_e = \frac{1}{2} C^{\alpha} (E^{\alpha})^2 + \frac{1}{2} C^{\gamma} [(E_1^{\gamma})^2 + (E_2^{\gamma})^2] \\ + \frac{1}{2} C^{\epsilon} [(E_1^{\epsilon})^2 + (E_2^{\epsilon})^2 + (E_3^{\epsilon})^2]$$

where

$$[3.35a] \quad C^{\alpha} = \frac{1}{3} (C_{11} + 2C_{12})$$

$$[3.35b] \quad C^{\gamma} = 2(C_{11} - C_{12})$$

$$[3.35c] \quad C^{\epsilon} = 4C_{44}$$

and

$$[3.36a] \quad E^{\alpha} = E_{xx} + E_{yy} + E_{zz}$$

$$[3.36b] \quad E_1^{\gamma} = \frac{(3)}{6} \frac{1}{2} (2E_{zz} - E_{xx} - E_{yy})$$

$$[3.36c] \quad E_2^{\gamma} = \frac{1}{2} (E_{xx} - E_{yy})$$

$$[3.36d] \quad E_1^{\epsilon} = E_{yz}$$

$$[3.36e] \quad E_2^{\epsilon} = E_{xz}$$

$$[3.36f] \quad E_3^{\epsilon} = E_{xy}$$

CHAPTER 4

MAGNETOELASTIC INTERACTIONS

The interactions between the magnetic moments on atoms in crystals depend upon the interatomic spacings. Therefore, the magnetic spin system is directly coupled to the atomic displacements. This coupling of the magnetic and elastic systems is called the magnetoelastic coupling. The total Hamiltonian of a magnetoelastic system consists of three parts - the magnetic Hamiltonian \mathcal{H}_m , the elastic Hamiltonian \mathcal{H}_e and the magnetoelastic coupling \mathcal{H}_{me} . We write the total Hamiltonian \mathcal{H} as:

$$[4.1] \quad \mathcal{H} = \mathcal{H}_m + \mathcal{H}_e + \mathcal{H}_{me} .$$

4.1 The Magnetoelastic Coupling \mathcal{H}_{me}

We have already discussed the magnetic Hamiltonian \mathcal{H}_m and the elastic Hamiltonian \mathcal{H}_e in previous chapters. We now discuss here the magnetoelastic coupling \mathcal{H}_{me} . Following Callen and Callen (1965) this may be formed by taking products of the symmetry strains and spin operators which transform according to the same irreducible representation

of the point group. In this way one can form an expression which is invariant under all the symmetry operations of the point group. The spin operators may involve spins on the same atom or spins on different atoms. We consider here only terms each of which consists of spin operators belonging to a single ion. These single-ion magnetoelastic terms may be written as

$$[4.2] \quad \mathcal{H}_{me}^I = - \sum_j \sum_{\Gamma} \sum_{\ell, m} \sum_{s, s'} M_{\ell m}^{\Gamma, s, s'} \sum_r E_r^{\Gamma, s}(j) \tilde{O}_{\ell m}^{\Gamma, r, s'}(S_j),$$

where Γ labels the irreducible representation and r labels the basis functions for that irreducible representation. s, s' label different sets of basis functions of Γ if it occurs more than once. The number of independent magnetoelastic coupling constants $M_{\ell m}^{\Gamma, s, s'}$ is determined by the particular point group. We have previously encountered the symmetry strains $E_r^{\Gamma, s}$ and the spin operators $\tilde{O}_{\ell m}^{\Gamma, s, s'}$.

For a cubic ferromagnetic crystal the single-ion magnetoelastic coupling has the form (Southern, 1973)

$$[4.3] \quad \mathcal{H}_{me}^I = \sum_j \mathcal{H}_m^I(j)$$

$$\begin{aligned}
[4.4] \quad \mathcal{H}_{me}^I(j) = & - M_0^\alpha E^\alpha - M_4^\alpha E^\alpha \{ \tilde{O}_{40}(\underline{S}_j) + (\frac{10}{7})^{\frac{1}{2}} \tilde{O}_{44}^+(\underline{S}_j) \} \\
& - M_2^Y \{ E_1^Y \tilde{O}_{20}(\underline{S}_j) + E_2^Y (2)^{\frac{1}{2}} \tilde{O}_{22}^+(\underline{S}_j) \} \\
& - M_4^Y \{ E_1^Y [\tilde{O}_{40}(\underline{S}_j) - (\frac{14}{5})^{\frac{1}{2}} \tilde{O}_{44}^+(\underline{S}_j)] \\
& + E_2^Y [- (\frac{24}{5})^{\frac{1}{2}} \tilde{O}_{42}^+(\underline{S}_j)] \} \\
& - M_2^E \{ E_1^E [i \tilde{O}_{21}^+(\underline{S}_j)] + E_2^E [- i \tilde{O}_{21}^-(\underline{S}_j)] \\
& + E_3^E [\tilde{O}_{22}^-(\underline{S}_j)] \} ,
\end{aligned}$$

where the symmetry strains are defined in Eqs. [3.36] and the operators $\tilde{O}_{\ell m}$, $\tilde{O}_{\ell m}^+$ and $\tilde{O}_{\ell m}^-$ are defined in Eqs. [2.21].

All the terms in \mathcal{H}_{me}^I are invariant under any of the symmetry operations of the point group. As an example we consider the third term in $\mathcal{H}_{me}^I(j)$. The constants M_0^α , M_4^α , M_2^Y , M_4^Y and M_2^E are called the magnetoelastic coupling constants.

We know from Eqs. [3.36b] and [3.36c] that

$$E_1^Y = \frac{(3)^{\frac{1}{2}}}{6} (2E_{zz} - E_{xx} - E_{yy})$$

$$E_2^Y = \frac{1}{2} (E_{xx} - E_{yy})$$

and from the tabulation of Buckmaster (1962) that

$$\begin{aligned}
[4.5] \quad \tilde{O}_{20}(\underline{s}_j) &= \frac{1}{2} [3s_z^2 - s(s+1)] \\
&= \frac{1}{2} [3s_z^2 - s^2] \\
&= \frac{1}{2} [3s_z^2 - (s_x^2 + s_y^2 + s_z^2)] \\
&= \frac{1}{2} [2s_z^2 - s_x^2 - s_y^2]
\end{aligned}$$

and

$$\begin{aligned}
[4.6] \quad \tilde{O}_{22}^+(\underline{s}_j) &= \frac{1}{2} [\tilde{O}_{22} + \tilde{O}_{2-2}] \\
&= \frac{1}{2} \cdot \frac{(6)}{4} \frac{1}{2} [s_+^2 + s_-^2] \\
&= \frac{1}{2} \cdot \frac{(6)}{4} \frac{1}{2} [(s_x + is_y)^2 + (s_x - is_y)^2] \\
&= \frac{(6)}{4} \frac{1}{2} [s_x^2 - s_y^2]
\end{aligned}$$

Therefore, the third term in $\mathcal{H}_{me}^I(j)$ is

$$\begin{aligned}
&= - \frac{(3)}{12} \frac{1}{2} M_2^Y \{ (2E_{zz} - E_{xx} - E_{yy}) (2s_z^2 - s_x^2 - s_y^2) \\
&\quad + 3(E_{xx} - E_{yy}) (s_x^2 - s_y^2) \}
\end{aligned}$$

Let us consider how this term transforms under a four-fold rotation of the coordinate system about the x-axis. Under

this transformation,

$$x \rightarrow x \quad \frac{\partial}{\partial x} \rightarrow \frac{\partial}{\partial x} \quad S_x \rightarrow S_x \quad \text{and} \quad u_x \rightarrow u_x$$

$$y \rightarrow -z \quad \frac{\partial}{\partial y} \rightarrow -\frac{\partial}{\partial z} \quad S_y \rightarrow -S_z \quad u_y \rightarrow -u_z$$

$$z \rightarrow y \quad \frac{\partial}{\partial z} \rightarrow \frac{\partial}{\partial y} \quad S_z \rightarrow S_y \quad u_z \rightarrow u_y$$

Then from Eq. [3.4]

$$E_{xx} \rightarrow E_{xx}$$

$$E_{yy} \rightarrow E_{zz}$$

$$E_{zz} \rightarrow E_{yy}$$

Therefore, the third term under this four-fold rotation of the coordinates, becomes

$$\begin{aligned} &= -\frac{(3)^{\frac{1}{2}}}{12} M_2^Y \{ (2E_{yy} - E_{xx} - E_{zz}) (2S_y^2 - S_x^2 - S_z^2) \\ &\quad + (E_{xx} - E_{zz}) (S_x^2 - S_z^2) \} \end{aligned}$$

After multiplying this out and rearranging the terms it may be seen that this is exactly the same as the original term in Eq. [4.4]. Therefore, this term is invariant under

this particular four-fold rotation. In the same way other symmetry operations may be shown to leave this expression invariant.

4.2 Magnetostriction

It is experimentally observed that a change in the magnetic state of a ferromagnetic or ferrimagnetic crystal is, in general, accompanied by a physical deformation of the crystal. This deformation is referred to as the spontaneous magnetostriction. The magnetostrictive strain which accompanies the magnetization to saturation of an elementary region of the body is anisotropic and depends on the orientation of the magnetization vector with respect to the crystallographic axes associated with this region. The observed bulk magnetostriction may be interpreted in terms of the spontaneous magnetostriction of each domain, but for simplicity we assume a single domain crystal. For the magnetizing field to be uniform throughout the crystal, the demagnetizing field should also be uniform throughout the crystal. This is possible only if the crystal is ellipsoidal in shape.

The deformation of the crystal does not arise wholly due to the spontaneous magnetization. There is an interaction, between the demagnetizing field with the spontaneous magnetization, which gives rise to an

additional lattice strain. This phenomenon is known as the form effect; but its contribution to the magnetostriction is usually negligibly small.

Let us suppose that in the absence of any magnetoelastic coupling an arbitrary point in the undeformed lattice has the coordinates x_i , and a neighboring point has the coordinates $x_i + \Delta x_i$ as in Fig. 4.1. When the magnetoelastic interaction is switched on, the first point is displaced by the vector \underline{u} and the second by $\underline{u} + \Delta \underline{u}$. Regarding u_i as a function of the independent variables x_j we can write

$$[4.7] \quad \Delta u_i = \frac{\partial u_i}{\partial x_j} \Delta x_j$$

We have already seen that the displacement gradient ($\partial u_i / \partial x_j$) can be written in terms of the symmetric strain function ϵ_{ij} and antisymmetric strain function ω_{ij} as

$$[4.8] \quad \frac{\partial u_i}{\partial x_j} = \epsilon_{ij} + \omega_{ij}$$

The antisymmetric strain function ω_{ij} , which represents a rotation of the elastic medium, must be zero if we are concerned only with equilibrium properties such as the magnetization and magnetostriction. However, in nonequilibrium situations (e.g., when a sound wave travels through a crystal) the ω_{ij} need not be zero.

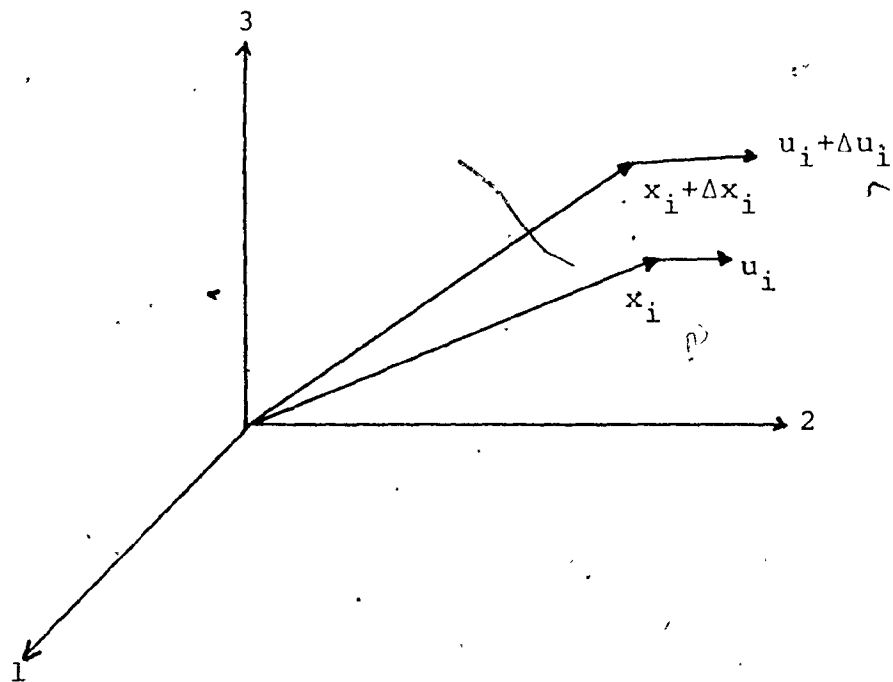


Fig. 4.1 Deformation of two neighbouring points.

Magnetostriction is measured by the fractional change in length of the elastic medium in some direction $\underline{\beta}$ specified by direction cosines $\beta_1, \beta_2, \beta_3$. We consider x_i and $x_i + \Delta x_i$ such that Δx_i is in the direction $\underline{\beta}$. If we define

$$[4.9] \quad \Delta x \equiv (\Delta x_j \Delta x_j)^{\frac{1}{2}}$$

then

$$\beta_1 = \frac{\Delta x_1}{\Delta x}, \quad \beta_2 = \frac{\Delta x_2}{\Delta x}, \quad \beta_3 = \frac{\Delta x_3}{\Delta x}$$

i.e., we can write

$$[4.10] \quad \beta_j = \frac{\Delta x_j}{\Delta x}$$

The change in the distance between two points after the magnetization is switched on is the vector Δu_j . The change in length along the direction $\underline{\beta}$ is

$$[4.11] \quad \begin{aligned} \delta l &= \Delta u_x \beta_1 + \Delta u_y \beta_2 + \Delta u_z \beta_3 \\ &= \beta_i \Delta u_i \end{aligned}$$

The fractional change in length measured in the direction $\underline{\beta}$ is

$$\begin{aligned}
 [4.12] \quad \lambda &= \frac{\delta l}{\Delta x} \\
 &= \frac{\beta_i \Delta u_i}{\Delta x} \\
 &= \beta_i \frac{\partial u_i}{\partial x_j} \frac{\Delta x_j}{\Delta x} \\
 &= \beta_i \beta_j \frac{\partial u_i}{\partial x_j}
 \end{aligned}$$

If the antisymmetric strain is zero then

$$[4.13] \quad \frac{\partial u_i}{\partial x_j} = \epsilon_{ij} ,$$

and therefore,

$$[4.14] \quad \lambda = \epsilon_{ij} \beta_i \beta_j .$$

In this derivation the magnetization has been assumed to be along some direction $\underline{\alpha}$ with direction cosines $\alpha_1, \alpha_2, \alpha_3$. Thus ϵ_{ij} is a function of $\underline{\alpha}$. We usually write λ_{100} as the fractional change in length measured along the (100) direction, and similarly for $\lambda_{110}, \lambda_{111}$, et cetera.

4.3 The Magnetoelastic Modes

In the presence of magnetoelastic coupling one no longer obtains purely magnetic or vibrational modes. The normal modes of the coupled system are mixed in character and are called magnetoelastic modes. They can be found by solving the equations of motion for the elastic displacements and the components of the magnetization which are a coupled system of differential equations.

In the present thesis we have adopted a microscopic description and the appropriate equations of motion involve the quantum mechanical spin operators and the phonon operators. The unperturbed magnon energies can be obtained by standard methods equivalent to the Holstein-Primakoff transformations. In what follows it is sufficient to consider only the lower magnon branch. In terms of the creation and destruction operators $\alpha_{\underline{q}}^{\dagger}$ and $\alpha_{\underline{q}}$ for magnons of wave vector \underline{q} in this lower branch, the magnetic Hamiltonian can be written in the form

$$[4.15] \quad \mathcal{H}_m = \sum_{\underline{q}} E_{\underline{q}} (\alpha_{\underline{q}}^{\dagger} \alpha_{\underline{q}} + \frac{1}{2}) ,$$

where $E_{\underline{q}}$ represents the magnon energy. The phonons are taken to have unperturbed $\hbar\omega_{\underline{q}\lambda}$, where λ labels the phonon branch. In terms of phonon creation and destruction operators $\beta_{\underline{q}\lambda}^{\dagger}$ and $\beta_{\underline{q}\lambda}$ the Hamiltonian for the lattice

vibrations can be written as

$$[4.16] \quad \mathcal{H}_e = \sum_{\mathbf{q}, \lambda} \hbar \omega_{\mathbf{q}\lambda} (\beta_{\mathbf{q}\lambda}^\dagger \beta_{\mathbf{q}\lambda} + \frac{1}{2}) .$$

In the last section the magnetoelastic coupling was described by a Hamiltonian consisting of products of strain functions and spin functions which satisfy the crystal symmetry. In order to express this coupling in microscopic terms, the spin operators must be expressed in terms of the $\alpha_{\mathbf{q}}^\dagger$ and $\alpha_{\mathbf{q}}$ using the Holstein-Primakoff transformation, and the strain functions must be expressed in terms of the $\beta_{\mathbf{q}\lambda}^\dagger$ and $\beta_{\mathbf{q}\lambda}$ using the well-known expansion for the displacement operators (see, for example, Kittel, 1963)

$$[4.17] \quad \underline{u}(\mathbf{j}) = \sum_{\mathbf{q}, \lambda} (2\rho\hbar\omega_{\mathbf{q}\lambda})^{-\frac{1}{2}} \hat{e}_{\mathbf{q}\lambda} (\beta_{\mathbf{q}\lambda} + \beta_{-\mathbf{q}\lambda}^\dagger) e^{i\mathbf{q}\cdot\mathbf{r}_j}$$

where ρ is the density of the medium and $\hat{e}_{\mathbf{q}\lambda}$ is a unit vector in the polarization direction. After a lengthy calculation the coupled Hamiltonian takes the form (Southern and Goodings, 1973)

$$[4.18] \quad \mathcal{H} = \sum_{\mathbf{q}} E_{\mathbf{q}} (\alpha_{\mathbf{q}}^\dagger \alpha_{\mathbf{q}} + \frac{1}{2}) + \sum_{\mathbf{q}, \lambda} \hbar \omega_{\mathbf{q}\lambda} (\beta_{\mathbf{q}\lambda}^\dagger \beta_{\mathbf{q}\lambda} + \frac{1}{2}) + \sum_{\mathbf{q}, \lambda} [V_{\mathbf{q}\lambda} \alpha_{\mathbf{q}} (\beta_{\mathbf{q}\lambda}^\dagger + \beta_{-\mathbf{q}\lambda}) + V_{\mathbf{q}\lambda}^* \alpha_{\mathbf{q}}^\dagger (\beta_{\mathbf{q}\lambda} + \beta_{-\mathbf{q}\lambda}^\dagger)]$$

where $V_{\underline{q}\lambda}$ is a complicated function of both the direction of magnetization and the applied magnetic field.

To find the coupled mode energies, we write down the equations of motion for the operators $\alpha_{\underline{q}}^\dagger$, $\alpha_{\underline{q}}$, $\beta_{\underline{q}\lambda}^\dagger$ and $\beta_{-\underline{q}\lambda}$, i.e.,

$$[4.19] \quad i\hbar \frac{\partial \alpha_{\underline{q}}^\dagger}{\partial t} = [\alpha_{\underline{q}}^\dagger, \mathcal{H}] \quad \text{et cetera.}$$

Now, if we consider only the coupling between the lower magnon branch and an impressed sound wave which has a definite direction of propagation and a definite polarization along one of the crystal axes, then by solving the equations of motion the coupled mode energies can be shown to be

$$[4.20] \quad \epsilon_{\underline{q}\lambda}^2 = \frac{1}{2} (E_{\underline{q}}^2 + \hbar^2 \omega_{\underline{q}\lambda}^2) \pm \frac{1}{2} [(E_{\underline{q}}^2 - \hbar^2 \omega_{\underline{q}\lambda}^2)^2 + 16E_{\underline{q}} \hbar \omega_{\underline{q}\lambda} |V_{\underline{q}\lambda}|^2]^{\frac{1}{2}}$$

We are usually concerned with behavior at long wavelengths, i.e., we have $\hbar \omega_{\underline{q}\lambda} \ll E_{\underline{q}}$. In this weak coupling limit the lower mode coupled energy is approximately given by

$$[4.21] \quad \epsilon_{\underline{q}\lambda}^2 = \hbar^2 \omega_{\underline{q}\lambda}^2 - \frac{4\hbar \omega_{\underline{q}\lambda} |V_{\underline{q}\lambda}|^2}{E_{\underline{q}}}$$

The elastic constant corresponding to a sound wave having a particular direction of propagation and a particular polarization is obtained from the sound velocity of the corresponding phonon mode in the long wavelength limit:

$$[4.22] \quad C = \rho V_s^2 = \rho \lim_{q \rightarrow 0} \left(\frac{\omega_{q\lambda}}{q} \right)^2 .$$

When the magnetoelastic coupling is included, the effective elastic constant C^* is obtained in a similar way from the energy of the appropriate magnetoelastic mode:

$$[4.23] \quad C^* = \rho \lim_{q \rightarrow 0} \left(\frac{\varepsilon_{q\lambda}}{\hbar q} \right)^2 .$$

Hence from Eq. [4.21] the expression for the effective elastic constant is

$$[4.24] \quad C^* = C - \lim_{q \rightarrow 0} \frac{4\rho\hbar\omega_{q\lambda} |v_{q\lambda}|^2}{E_q (\hbar q)^2} .$$

The expressions for $v_{q\lambda}$ and E_q in Eq. [4.24] are very much simplified if both the direction of magnetization and applied magnetic field are along one of the cubic symmetry directions. The full expressions for $v_{q\lambda}$ for the various possible directions of q and λ (the polarization) are given in the appendix of Southern and Goodings (1973) and will not be repeated here.

4.4 Elastic Constants in Terms of Macroscopic Quantities

The relation for the effective elastic constant in Eq. [4.24] is not usually used for actual calculations. For practical purposes this relation is written in terms of macroscopic quantities which could be derived easily from the free energy of the magnetic system. Southern (1973a) has shown that, when both the direction of magnetization and the applied magnetic field coincide with one of the crystal symmetry axes, the transverse elastic waves which are either propagating or polarized in the magnetization direction can always be expressed in the form,

$$[4.25] \quad C_T^* = C_T - \frac{(MH_{ME} \pm MH_A)^2}{4M(H_A + H)},$$

where the upper sign refers to a transverse wave propagating in the magnetization direction and the lower sign refers to a transverse wave polarized in this direction, H is the applied magnetic field and M is the magnetization. Both the effective magnetoelastic field H_{ME} and the effective anisotropy field H_A depend on the direction of the magnetization and the plane defined by the propagation and the polarization directions of the elastic wave.

To eliminate the domain alignment effects we assume that some minimum field strength H_0 is required to

achieve saturation of the magnetization and we write the Eq. [4.25] in the form

$$[4.26] \quad C^*(H) - C^*(H_0) = \frac{M(H-H_0) [H_{ME} \pm H_A]^2}{4(H_A+H)(H_A+H_0)}$$

The expressions for H_{ME} and H_A can be obtained from the free energy. Both H_{ME} and H_A not only depend on the direction of magnetization, they also depend on which elastic constant is being measured. In Table 4.1 we quote the expressions for MH_{ME} and MH_A obtained by Southern (1973) for a cubic crystal. The effective anisotropy field MH_A is given in terms of the usual macroscopic cubic anisotropy constants K_1 and K_2 which are related to the microscopic constants defined in the magnetic Hamiltonian in Eq. [2.23] as follows

$$[4.27a] \quad K_1 = -\frac{1}{2} [10b_4 \hat{I}_{9/2} + 21b_6 \hat{I}_{13/2}]$$

$$[4.27b] \quad K_2 = \frac{231}{2} b_6 \hat{I}_{13/2}$$

where

$$[4.28a] \quad b_4 = \tilde{B}_4^0 N_a SS\left(\frac{3}{2}\right)$$

$$[4.28b] \quad b_6 = \tilde{B}_6^0 N_a SS\left(\frac{5}{2}\right)$$

TABLE 4.1

EXPRESSIONS FOR MH_{ME} AND MH_A

Direction of Magnetization	Elastic Constant Measured	MH_A	MH_{ME}
[001]	C_{44}	$2K_1$	$-2\beta_2 + \beta_5$
[111]	$\frac{1}{3}(C_{11}-C_{12}+C_{44})$	$-\frac{4}{3}K_1 - \frac{4}{9}K_2$	$-\frac{4}{3}\beta_1 - \frac{2}{3}\beta_2 + \frac{16}{63}\beta_4 + \frac{10}{9}\beta_5$
[110]	$\frac{1}{2}(C_{11}-C_{12})$	$-2K_1$	$-2\beta_1 - \frac{2}{7}\beta_4$
[110]	C_{44}	$K_1 + \frac{1}{2}K_2$	$-2\beta_2 - \frac{5}{2}\beta_5$

$$[4.29] \quad s(n) = (S - \frac{1}{2})(S-1)(S - \frac{3}{2}) \dots (S-n)$$

and N_a is the number of magnetic atoms per unit volume. The temperature dependence is taken into account in the reduced hyperbolic Bessel function $\hat{I}_{\ell+1/2}(\mathcal{L}^{-1}(m))$ where $\mathcal{L}^{-1}(m)$ is the inverse Langevin function and m is the reduced magnetization. The effective magnetoelastic field MH_{ME} is given in terms of the magnetoelastic constants of Kittel and Van Vleck (1960) which are related to those defined in the coupled magnetoelastic Hamiltonian in Eq. [4.4] as follows:

$$[4.30a] \quad \beta_0 = - M_0^\alpha N_a$$

$$[4.30b] \quad \beta_1 = - \left(\frac{3}{4}\right)^{\frac{1}{2}} M_2^Y N_a SS\left(\frac{1}{2}\right) \hat{I}_{5/2}$$

$$[4.30c] \quad \beta_2 = - \left(\frac{3}{8}\right)^{\frac{1}{2}} M_2^\epsilon N_a SS\left(\frac{1}{2}\right) \hat{I}_{5/2}$$

$$[4.30d] \quad \beta_3 = 5M_4^\alpha N_a SS\left(\frac{3}{2}\right) \hat{I}_{9/2} - \frac{7(3)^{\frac{1}{2}}}{3} M_4^Y N_a SS\left(\frac{3}{2}\right) \hat{I}_{9/2}$$

$$[4.30e] \quad \beta_4 = - \frac{7(3)^{\frac{1}{2}}}{2} M_4^Y N_a SS\left(\frac{3}{2}\right) \hat{I}_{9/2}$$

$$[4.30f] \quad \beta_5 = (5)^{\frac{1}{2}} M_4^\epsilon N_a SS\left(\frac{3}{2}\right) \hat{I}_{9/2}$$

CHAPTER 5

NUMERICAL ESTIMATES FOR EUROPIUM CHALCOGENIDES

In the previous chapters we have described the theory of magnetoelastic interactions for cubic crystals. In this chapter we estimate magnetoelastic effects in the europium chalcogenide crystals. The basic data for these materials are listed in Table 5.1; the elastic constants in Table 5.2, the experimental values of anisotropy constants and the estimated crystal field parameters in Table 5.3, and the experimental values of magnetostriction coefficients, the estimated values of magnetostriction constants and the magnetoelastic constants in Table 5.4. The data for the anisotropy constants and magnetostriction coefficients is complete only for europium oxide. Therefore, our estimates of the field dependence of the elastic constants will be limited to europium oxide.

5.1 Estimates of Crystal Field Parameters and Magnetoelastic Constants for EuO

Measurements of the anisotropy constants of EuO as a function of temperature have been reported by Hughes,

TABLE 5.1

BASIC DATA FOR EUROPIUM CHALCOGENIDES

(McGuire and Shafer, 1964)

Compound	Lattice Parameters Å	No. of Eu ions per cm ³ $N_a (\times 10^{22})$	Bohr Magnetons Per Eu ion $\mu_B (T=0)$	Saturation Magnetization In Gauss M (T=0)	Critical Temperature $T_C (^\circ K)$
EuO	5.14	2.95	6.80	1860	69
EuS	5.96	1.89	6.87	1204	16.5
EuSe	6.19	1.69	6.70	1050	7
EuTe	6.60	1.40	-	-	7.8

TABLE 5.2
ELASTIC CONSTANTS (Shapira and Reed, 1971)

Compound	C_{11} (in 10^{11} CGS unit)	C_{44} (in 10^{11} CGS unit)	C_{12} (in 10^{11} CGS unit)
EuO	19.2 ± 0.6	5.42 ± 0.13	4.25 ± 0.85
EuS	13.1 ± 0.5	2.73 ± 0.11	1.1 ± 0.8
EuSe	11.6 ± 0.4	2.28 ± 0.09	1.2 ± 0.6
EuTe	9.36 ± 0.4	1.63 ± 0.07	0.67 ± 0.6

TABLE 5.3

ANISOTROPY CONSTANTS AND CRYSTAL FIELD PARAMETERS

Compound	K_1/M (Gauss)	K_2/M (Gauss)	References	K_1 ergs/cm ³	K_2 ergs/cm ³	b_4 ergs/cm ³	b_6 ergs/cm ³
EuO	-240	70	Hughes et al., 1974	-4.46×10^5	1.30×10^5	8.68×10^3	1.1×10^3
EuS	-19.6	-4.6	Brown et al., 1968	-	-	-	-
EuSe	-50	-83	do	-	-	-	-

TABLE 5.4

MAGNETOSTRICTION COEFFICIENTS, MAGNETOSTRICTION CONSTANTS
AND MAGNETOELASTIC CONSTANTS

Compound	T = 0°K	References	β_1 ergs/cm ³	β_2 ergs/cm ³	M_2^Y ergs/ions	M_2^E ergs/ions	
	λ_{100}						
	λ_{111}						
EuO	-22×10^{-6}	55×10^{-6}	Argyle et al., 1968	3.58×10^7	-8.94×10^7	-1.33×10^{-16}	4.71×10^{-16}

1968

Everett and Lawson (1974). Their results show the temperature dependence expected for single ion anisotropy originating in the crystal field. From their results of K_1/M and K_2/M listed in Table 5.3 and using the value of M in Table 5.1 we can find the values of K_1 and K_2 , at $T=0$, which are listed in Table 5.3. From these values we can now estimate the crystal field parameters b_4 and b_6 defined in Eqs. [4.28]. At $T=0$ the reduced hyperbolic Bessel functions $\hat{I}_{\ell+1/2}(m)$ in Eqs. [4.27] become equal to unity and the relations between $K_1(0)$, $K_2(0)$ and b_4 , b_6 are

$$[5.1] \quad K_1 = -5b_4 - \frac{21}{2} b_6$$

$$[5.2] \quad K_2 = \frac{231}{2} b_6$$

Now solving these two equations we obtain the values of b_4 and b_6 listed in Table 5.3. These values are in agreement with those quoted by Hughes et al. (1974) when differences in notation are taken into account. By comparing these estimates with the values of b_4 and b_6 quoted by Southern (1973) for the rare earth iron cubic Laves phase compounds we find that they are more than a factor of 10^3 smaller.

The magnetostriction coefficients of EuO single crystal at 0°K were measured by Argyle and Miyata (1968). The values of λ_{100} and λ_{111} are listed in Table 5.4. These two measurements alone are insufficient to obtain numerical

values for all the independent magnetostriction constants. Therefore, following Southern (1973), we assume that β_1 and β_2 are the most important and that the other constants are negligibly small. From Eqs. (3.3a) and (3.3b) of Southern (1973) we have

$$[5.3] \quad \beta_1 = -3\mu\lambda_{100}$$

$$[5.4] \quad \beta_2 = -3\mu\lambda_{111}$$

where μ is the shear modulus which can be obtained from the relation (see, for example, Cottrell, 1964)

$$[5.5] \quad \mu = C_{44}$$

Using the value of C_{44} listed in Table 5.2 and from Eqs. [5.3], [5.4] and [5.5], we obtain the values of β_1 and β_2 which are listed in Table 5.4. Then the magnetoelastic constants M_2^Y and M_2^E are obtained from their relations of β_1 and β_2 in Eqs. [4.30b] and [4.30c]; these values are listed in Table 5.4. If we compare again these values of β_1 and β_2 with those of the rare earth iron cubic Laves phase compounds listed in Southern (1973) we find that they are more than a factor of 10^2 smaller.

5.2 Changes in Elastic Constants due to Magnetoelastic Interaction

Now using the values of K_1 , K_2 , β_1 and β_2 given in Tables 5.3 and 5.4, we can calculate the values of MH_A and MH_{ME} in Table 4.1 for the various elastic constants measured for different directions of the magnetization. These values are listed in Table 5.5.

In Figs. 5.1 to 5.4 we have plotted $[\bar{C}^*(H) - C^*(H_0)]/C^*$ as a function of $(H-H_0)/H_0$. It can be easily seen from the figures that the fractional change in the elastic constant C_{44} , when the magnetic field is along [001] or [110] direction and the transverse wave is either propagating or polarized in the direction of the magnetic field, is as large as 5×10^{-4} for an applied field up to 25 kOe. The fractional change for $\frac{1}{2}(C_{11}-C_{12})$, when the magnetic field is along [110], is of the order of 10^{-5} , and the fractional change for $\frac{1}{3}(C_{11}-C_{12}+C_{44})$, when the magnetic field is along [111], is of the order of 10^{-6} .

Melcher (1970) has reported the measurements of $\frac{\Delta C}{C}$ of the order of 10^{-6} for antiferromagnetic MnF_2 . But up to the present time no measurements have been reported for any ferromagnetic materials which would verify the predictions of the finite strain theory. The reason for this is mainly the incompatible requirements on sample shape for performing measurements of the sound velocity in

Fig. 5:1 $\Delta C_{44}/C_{44}$ as a function of $(H-H_0)/H_0$ for EuO with the magnetic field along [001] and $H_0 = 10$ kOe. Solid (dashed) curves are for propagation (polarization) along [001].

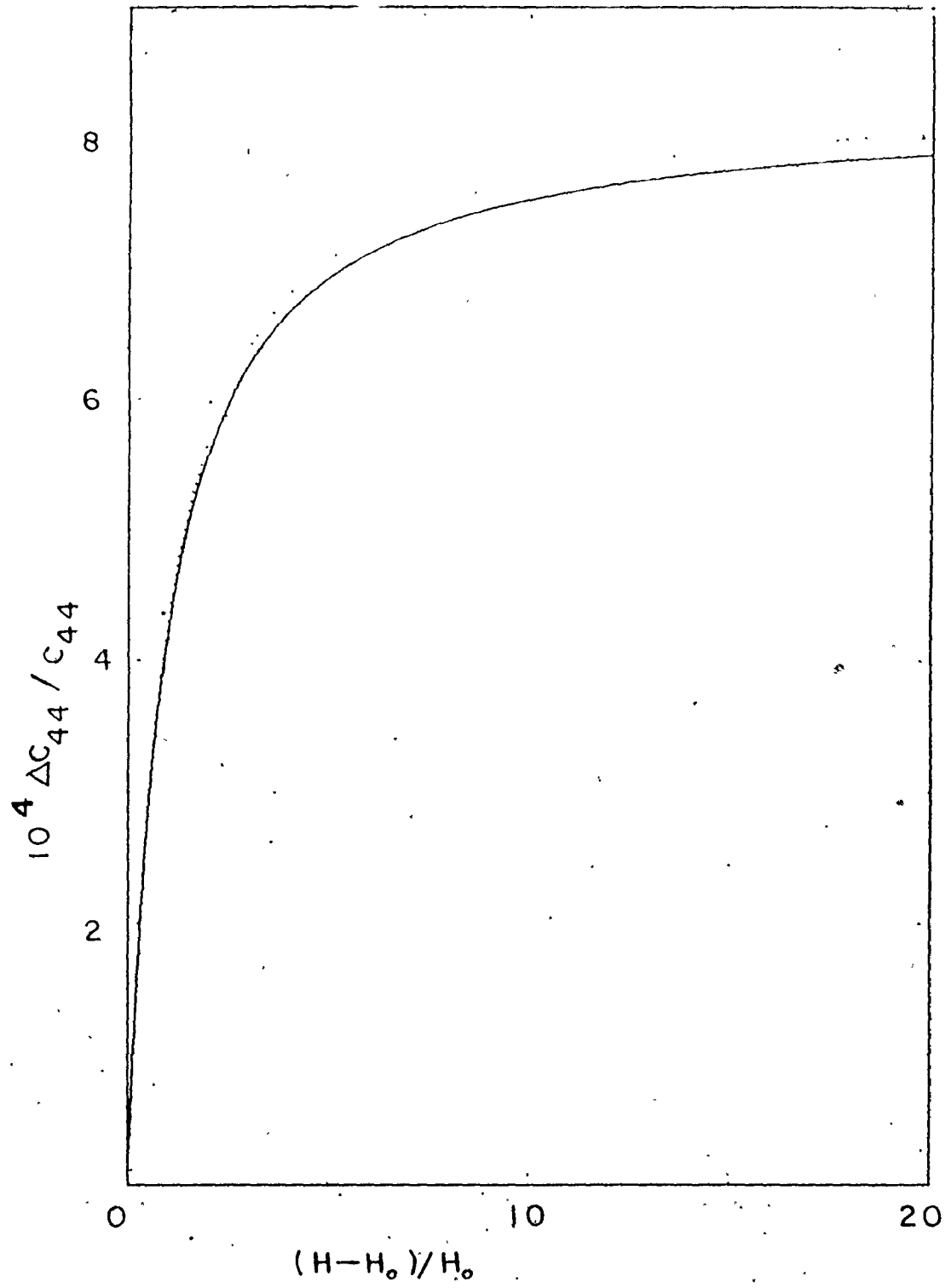


Fig. 5.1

Fig. 5.2 $\Delta C/C$ as a function of $(H-H_0)/H_0$ for EuO with the magnetic field along [111] and $H_0 = 10$ kOe. Solid (dashed) curves are for propagation (polarization) along [111] and $C = \frac{1}{3} (C_{11} - C_{12} + C_{44})$.

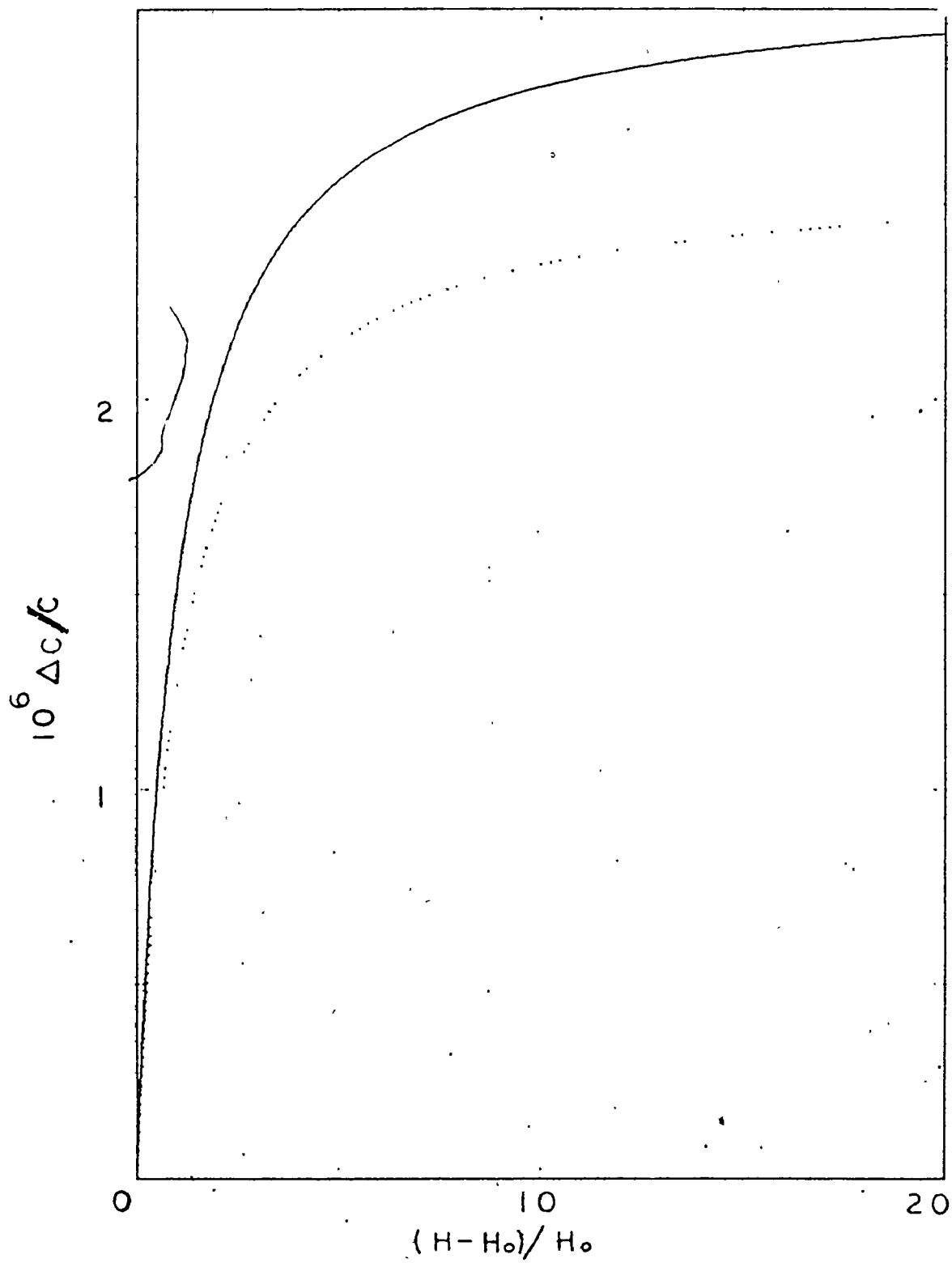


Fig. 5.2

Fig. 5.3 $\Delta C/C$ as a function of $(H-H_0)/H_0$ for EuO with the magnetic field along $[11\bar{0}]$ and $H_0 = 10$ kOe. Solid (dashed) curves are for propagation (polarization) along $[110]$ and $C = \frac{1}{2} (C_{11}-C_{12})$.

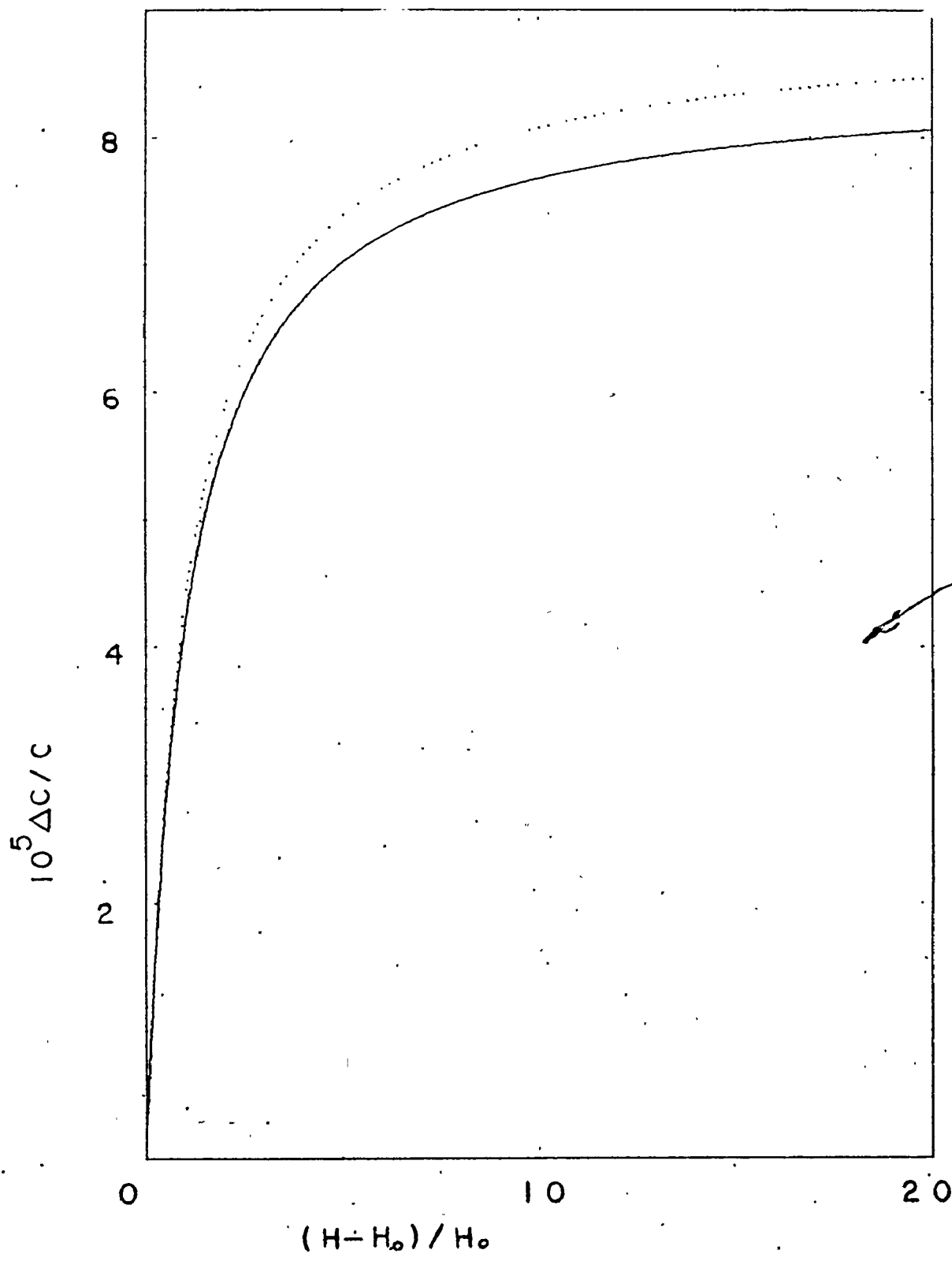


Fig. 5.3

Fig. 5.4 $\Delta C_{44}/C_{44}$ as a function of $(H-H_0)/H_0$ for EuO with
the magnetic field along [110] and $H_0 = 10$ kOe.
Solid (dashed) curves are for propagation
(polarization) along [110].

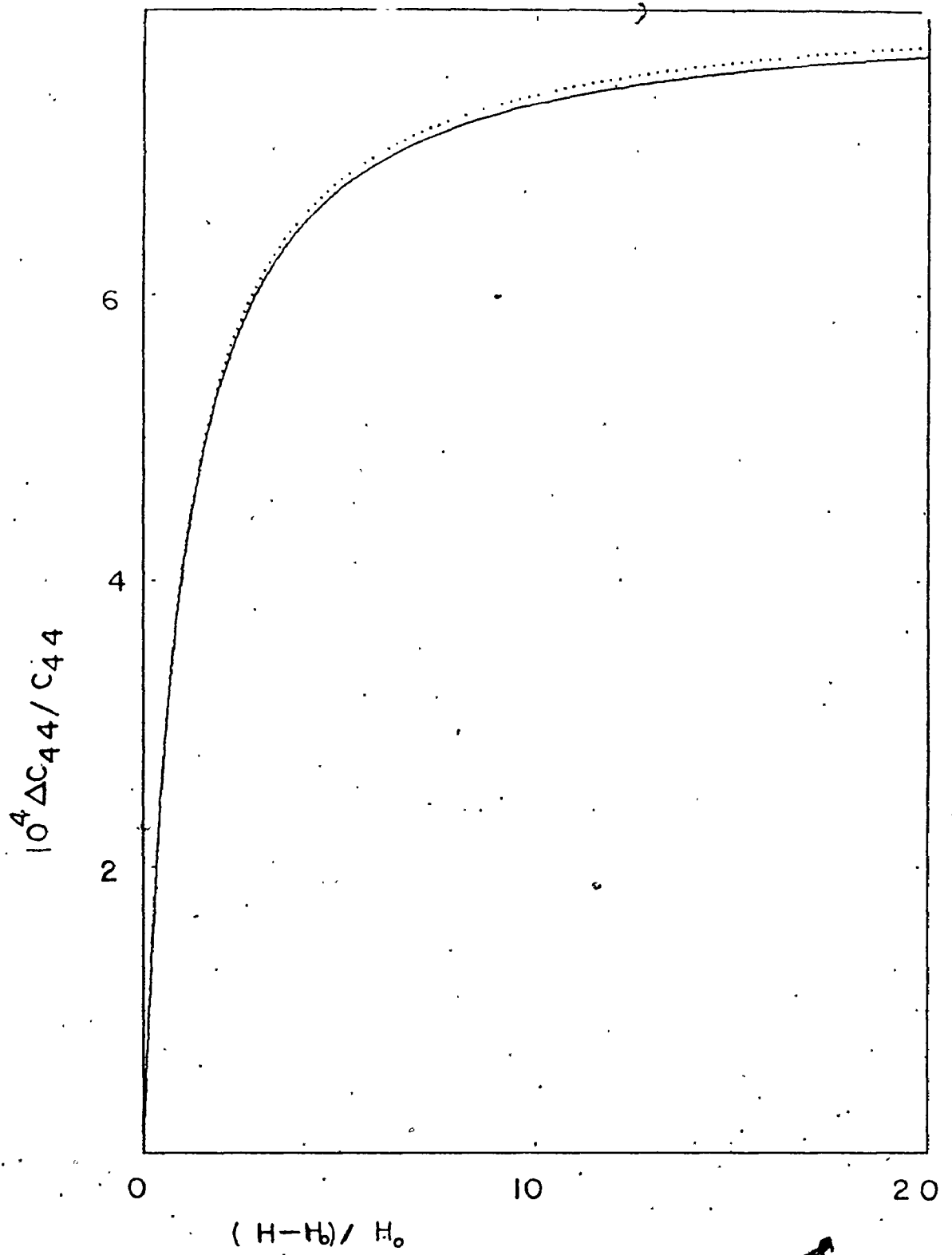


Fig. 5.4

a ferrimagnetic crystal. On the one hand it is desirable to have the crystal shaped in the form of an ellipsoid in order that the magnetization be uniform throughout the crystal. However, sound velocity measurements by the phase echo technique require two parallel surfaces. A sample in the form of a disk satisfies both of these requirements and it should be possible to grow good single crystals having this shape. We, therefore, believe that it will be possible to perform measurements on EuO corresponding to the calculations reported in this thesis. On the basis of our calculations, the largest effect can be expected for measurements of the elastic constant C_{44} .

CHAPTER 6

SUMMARY

The theory of magnetoelastic interactions has been discussed for ferromagnetic crystals with cubic symmetry in this thesis. A magnetic system is described where the energy due to magnetoelastic anisotropy is included; and an elastic system is described in the framework of finite strain theory. The coupling of the magnetic and the elastic systems is taken into account through single ion interactions. The coupled modes of such a magnetoelastic system are discussed, and expressions for the changes in various elastic constants as a function of an applied magnetic field are written down.

Using the available data for anisotropy constants and magnetostriction coefficients we have estimated the crystal field parameters and the magnetoelastic constants for EuO . It is predicted that the fractional change in the measured elastic constant C_{44} is of the order of 5×10^{-4} in an applied field of 25 kOe. Furthermore, the difference between the values of C_{44} measured for sound waves propagating along the [001] or [110] directions and polarized in the other direction is calculated to be of the

order of 10^{-5} . It may be possible, by analyzing the measurements of the fractional change in the various elastic constants and their difference for two types of transverse wave, to estimate the anisotropy constants and magnetoelastic constants.

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