MAGNETOELASTIC EFFECTS IN RARE EARTH METALS

AND COMPOUNDS

MAGNETOELASTIC EFFECTS IN RARE EARTH METALS AND COMPOUNDS

By

BYRON WAYNE SOUTHERN, B.SC., M.SC.

A Thesis

Submitted to the School of Graduate Studies

in Partial Fulfilment of the Requirements

for the Degree

Doctor of Philosophy

McMaster University

June 1973

DOCTOR OF PHILOSOPHY (1973) McMASTER UNIVERSITY
(Physics) Hamilton, Ontario.

TITLE: Magnetoelastic Effects in Rare Earth Metals and Compounds

AUTHOR: Byron Wayne Southern, B.Sc. (York University)
M.Sc. (McMaster University)

SUPERVISOR: Professor D. A. Goodings

NUMBER OF PAGES: (vii), 135

SCOPE AND CONTENTS:

A theory developed by Toupin, Tiersten, Brown and Melcher employing finite strains and angular momentum invariants is applied to ferromagnets which have hexagonal-close-packed or cubic symmetry. A Hamiltonian is written down which includes Heisenberg-exchange, magnetic anisotropy, magnetoelastic terms and the Zeeman term and which is invariant under combined rotations of the magnetic and elastic systems. When the approximations of small-strain theory are subsequently carried out, there appear new terms originating in the magnetic anisotropy that are linear in the antisymmetric strains $\omega_{\mu\nu}$ and correspond to rotations of the elastic medium. The coupling of transverse acoustic waves to the magnetic system is studied and expressions are derived for the dependence of the measured elastic constants

on an applied magnetic field in the ferromagnetic phase. Using available data on magnetic anisotropy and magnetostriction, estimates are given for the size of the effects that may be expected to be found in the rare-earth metals (Gd, Tb, Dy, Ho and Er) and in some of the rare-earth-iron compounds RFe_2 (R = Tb, Dy, Ho, Er and Tm). Fractional changes in the elastic constants as large as 10^{-2} are predicted in fields of about 50 kOe. Calculations are also performed for the field-dependent changes in c_{11} and c_{33} for longitudinal waves in the paramagnetic region for Dy and Ho.

ACKNOWLEDGEMENTS

I wish to thank my research supervisor, Dr. D. A. Goodings, for his valuable suggestions and assistance in this investigation.

A discussion with Dr. J. Grindlay and correspondence with Dr. R. L. Melcher concerning rotational invariance in coupled magnetic and elastic systems is gratefully acknowledged.

I would also like to thank Dr. A. E. Clark and Dr. M. P. Dariel for sending me unpublished results obtained with their colleagues.

Financial support from the National Research Council of Canada is also gratefully acknowledged.

Many thanks to Miss Erie Long for the fast and accurate typing of this thesis.

TABLE OF CONTENTS

TABLE OF CONTENTS - continued

TABLE OF CONTENTS - continued

CHAPTER 1

INTRODUCTION

1.1 Magnetic Interactions

The magnetic moments on atoms in solids may interact with each other in a number of ways. However, the simplest model of magnetism is one in which electrons in well-localized magnetic d- or f-shells interact with one another via an exchange mechanism having the Heisenberg form $-\sum_{i<j} J_{ij}\, \underset{\sim}{S}_i \cdot \underset{\sim}{S}_j$. Here $\underset{\sim}{S}_i$ represents the total spin angular momentum of the i^{th} atom and J_{ij} denotes the exchange integral which depends upon the separation distance of the atoms. The low energy excitations of spin systems coupled by exchange interactions are wave-like and are called spin waves. In a spin wave, a small sinusoidally varying deviation of the moments from the completely ordered state travels through the lattice. The energy of a spin wave is quantized and these quantized excitations are referred to as magnons. The excitation of a single magnon reduces the ordered moment of the system by one Bohr magneton. If the interactions between magnons are neglected, then they have a well defined energy $E_{\underset{\sim}{q}}$ and momentum $\hbar\underset{\sim}{q}$. Classically, these excitations correspond to a precession of the individual

magnetic moments about the ordered moment direction and the phase of one moment relative to another is determined by the wavevector $\underline{q}$. Exchange forces are mainly responsible for determining the dispersion relation $E_{\underline{q}}$ vs $\underline{q}$ and hence a determination of the dispersion curve by neutron inelastic scattering techniques will give information about the exchange interactions.

The exchange forces, however, do not give rise to any preferred orientation of the ordered moment with respect to the crystal axes. In a ferromagnetic crystal it is generally found that the direction of the ordered moment coincides with certain definite crystallographic axes. These axes are referred to as directions of easy magnetization or magnetically easy directions. All other symmetry directions are called magnetically hard directions. We can say that there are effective internal magnetic fields in the crystal which tend to orient the magnetic moments along certain preferred axes. The energy of the magnetic spin system depends on the orientation of the magnetization with respect to the crystal axes and we refer to this energy as the magnetocrystalline anisotropy energy.

Since the interactions between the magnetic moments on atoms in crystals depend upon the interatomic spacings, the magnetic spin system is directly coupled to the atomic displacements. This coupling of the magnetic and elastic systems is called magnetoelastic coupling and it is comprised

of both a static and a dynamic part. In a ferromagnetic crystal the static part of this coupling results in a shift of the equilibrium positions of atoms from those they would occupy if there were no coupling. The crystal symmetry is slightly distorted compared to the paramagnetic state and the distortion depends upon the direction of the spontaneous magnetization. This static effect is usually referred to as magnetostriction and it contributes to the effective anisotropy field in determining the equilibrium direction of the ordered moment. The dynamic part of the magnetoelastic interaction couples together the oscillations of both the magnetic moments and the elastic displacements about the equilibrium configuration of the combined system. These oscillations in the magnetic and elastic degrees of freedom are usually described in terms of magnons and phonons respectively. Phonons describe the quantized lattice vibrations in the same way that magnons describe the quantized spin deviations. The low energy excitations of the combined magnetic and elastic system are only adequately described in terms of these purely magnetic and purely elastic normal modes if the coupling between them is sufficiently weak. If the dynamic interaction is not weak, then the correct normal modes of the system possess both magnon and phonon characteristics and should be referred to as magnetoelastic modes.

In certain materials, magnetoelastic interactions strongly influence both the equilibrium and nonequilibrium magnetic properties. As mentioned previously, the static magnetoelastic coupling contributes to the effective anisotropy field in the crystal. In addition to affecting the equilibrium moment direction, the contributions to this effective anisotropy field also influence the magnetic excitations or spin waves. These effects may be investigated experimentally using ferromagnetic resonance techniques.

In a ferromagnetic resonance absorption experiment, a static magnetic field is applied in the magnetization direction and a small oscillating magnetic field is applied perpendicularly to this static field. The static field strength, together with the effective anisotropy field, determine the precession frequency of the magnetic moments. When this precession frequency matches the frequency of the oscillating field, an absorption of energy from the field can be observed. By measuring the value of the static field required for resonance as a function of temperature, information about the effective anisotropy field may be obtained.

In addition to having a strong influence on the static magnetic properties, the magnetoelastic coupling also has interesting effects on the measurement of the dynamic elastic properties. In particular, the measured elastic constant, or equivalently the sound velocity, depends on the presence of magnetic order and also upon the application of

a magnetic field. Measurements of this type have been used to obtain information about the magnetoelastic coupling constants (LeCraw and Kasuya 1963). These values may then be compared with those obtained in magnetostriction measurements using standard strain-gauge techniques.

1.2 Earlier Treatments of Dynamic Magnetoelastic Coupling

The earliest treatments of dynamic magnetoelastic coupling (Kittel 1958; Akhiezer et al. 1961) are formulated in the continuum limit where the wavelengths of both the spin waves and the elastic waves are long compared to the interatomic spacings. Macroscopic variables describing the magnetic and elastic degrees of freedom are defined and the laws of continuum physics are applied to obtain the classical equations which govern the behaviour of the system. In addition, an energy density is formed to describe the magnetic and elastic energies as well as the magnetostrictive energy. However, this function is formed under the assumption that the results of classical elasticity theory remain valid in the coupled magnetoelastic medium. Both Tiersten (1964) and Brown (1964) have demonstrated that this assumption leads to a violation of the law of conservation of total angular momentum in the medium.

In the usual theory of elasticity (see, for example, Love 1944), the laws of conservation of mass, linear momentum, angular momentum and energy are applied to an elastic medium. The conservation of mass and linear momentum leads directly to the usual equations of motion for the elastic displacements

$$[1.1] \quad \rho \ddot{\underline{u}} = \underline{f}$$

where ρ is the density of the medium and $\underline{f}$ is the volume force density. This force is usually expressed as the gradient of the mechanical stress tensor t_{ij} as follows

$$[1.2] \quad f_i = t_{ij,j} \equiv \frac{\partial t_{ij}}{\partial x_j} \qquad (i = 1, 2, 3)$$

where repeated indices are to be summed over. Moreover, a Hooke's law relation is usually assumed to exist between stress and strain in the medium. The application of the law of conservation of energy allows the stress tensor to be expressed as the gradient of an elastic energy density with respect to the infinitesimal strain tensor ϵ_{ij} as follows

$$[1.3] \quad t_{ij} = \frac{\partial F^{(e)}}{\partial \epsilon_{ij}} \quad .$$

The elastic energy density $F^{(e)}$ is usually written as

$$[1.4] \qquad F^{(e)} = \frac{1}{2} c_{ijk\ell}\, \varepsilon_{ij} \varepsilon_{k\ell}$$

where, as usual, summation over repeated indices is implied and the number of independent elastic constants $c_{ijk\ell}$ is determined by the point group symmetry of the medium. Combining [1.2] and [1.3], the equations of motion for the elastic displacements may be rewritten in the form

$$[1.5] \qquad \rho \ddot{u}_i = \frac{\partial^2 F^{(e)}}{\partial x_j \partial \varepsilon_{ij}} \qquad (i = 1, 2, 3) \quad .$$

The usual theory of elasticity also applies the law of conservation of angular momentum to the elastic medium and obtains the result that the mechanical stress tensor is symmetric. This fact has already been assumed in eq. [1.3] since the elastic energy density $F^{(e)}$ only involves the symmetric strain components ε_{ij}. Thus, the form of the equations of motion for the elastic displacements [1.5] is valid provided the energy density depends only on the symmetric strains ε_{ij}.

In a magnetically ordered material, the magnetostrictive energy density is usually written in the following form (Becker and Döring 1939; Kittel 1958)

$$[1.6] \qquad F^{(me)} = b_{ijk\ell}\, \alpha_i \alpha_j\, \varepsilon_{k\ell}$$

where $\alpha_i (i = 1, 2, 3)$ are the direction cosines of the

spontaneous magnetization. As in the case of the elastic constants, the number of independent magnetoelastic coupling constants $b_{ijk\ell}$ is determined by the point group symmetry. Additional terms involving higher powers of the direction cosines α_i may also be included. This form of the magnetostrictive energy describes the static coupling of the magnetic and elastic systems. As mentioned previously, several authors have used the same form of the magnetostrictive energy to examine the dynamic magnetoelastic coupling. Tiersten (1964) and Brown (1964) have argued that this approach corresponds to applying the law of conservation of angular momentum to the magnetic and elastic systems separately.

It is a well known fact that a magnetic moment possesses angular momentum and that a magnetic field acting on a magnetic moment produces a couple equal to $\underset{\sim}{M}\times\underset{\sim}{H}$. Such couples are taken to be zero in the usual treatments of elasticity theory. The application of the law of conservation of angular momentum to the magnetic system alone yields the usual torque equations

$$[1.7] \qquad \frac{d\underset{\sim}{M}}{dt} = \gamma(\underset{\sim}{M}\times\underset{\sim}{H}^{eff})$$

where γ is the gyromagnetic ratio. This equation assumes that the magnitude of the magnetic moment remains constant

and that only its direction may change. The effective magnetic field $\underset{\sim}{H}^{eff}$ acting on the magnetic moment may be expressed in terms of a magnetic energy density using the law of conservation of energy in the magnetic system. We have (Akhiezer et al. 1961)

$$[1.8] \quad \underset{\sim}{H}^{eff} = - \frac{\partial F^{(m)}}{\partial \underset{\sim}{M}}$$

where the energy density $F^{(m)}$ includes the effects of exchange, magnetic anisotropy and magnetic dipole interactions. This energy density $F^{(m)}$ may be expanded in a power series involving even powers of the direction cosines α_i as follows

$$[1.9] \quad F^{(m)} = K_{ij}\alpha_i\alpha_j + K_{ijk\ell}\alpha_i\alpha_j\alpha_k\alpha_\ell + \cdots$$

The coefficients K_{ij} and $K_{ijk\ell}$ are called magnetic anisotropy coefficients and the point group symmetry determines the number of independent coefficients that are required. Combining [1.7] and [1.8], the equations of motion for the magnetization components may be written as

$$[1.10] \quad \frac{d\underset{\sim}{M}}{dt} = \gamma [\underset{\sim}{M} \times (- \frac{\partial F^{(m)}}{\partial \underset{\sim}{M}})] \quad .$$

The standard treatments of dynamic magnetoelastic coupling correspond to taking the total energy density F for the magnetoelastic medium to be the sum of the energy

densities in [1.4], [1.6] and [1.9]. The equations of motion [1.5] and [1.10] for the elastic displacements and the components of magnetization respectively are then written down with both $F^{(e)}$ and $F^{(m)}$ replaced by F. The solutions of these coupled equations involve the various coupling constants defined in $F^{(e)}$, $F^{(me)}$ and $F^{(m)}$ and, consequently, depend strongly on the actual form chosen for the total energy density. However, as mentioned previously, this approach corresponds to the application of the law of conservation of angular momentum to the two systems separately. If the couples which are taken to be zero in the usual theories of elasticity are included, then the application of the law of conservation of total (spin plus lattice) angular momentum in the combined magnetic and elastic system yields the result that the stress tensor is no longer symmetric. In fact, the antisymmetric portion of the stress tensor is related to the total energy density F as follows (Melcher 1971)

$$[1.11] \quad t_{ij} - t_{ji} = M_i \frac{\partial F}{\partial M_j} - M_j \frac{\partial F}{\partial M_i} \ .$$

The construction of a total energy density to describe the combined magnetic and elastic systems must be consistent with [1.11], otherwise such a function clearly violates the law of conservation of angular momentum.

One method of ensuring that this law is not violated is to construct the energy function, or Hamiltonian, such that it is invariant with respect to arbitrary rotations of the combined magnetoelastic medium. This concept of rotational invariance was first introduced by Toupin (1956) in his treatment of the elastic dielectric. Tiersten (1964) and Brown (1966) later applied the same idea to a magnetoelastic medium. However, a clear understanding of this concept requires certain of the ideas of finite strain theory and these are discussed in detail in the next chapter.

We shall see later that the standard treatments of dynamic magnetoelastic coupling in magnetically anisotropic media are incorrect. These treatments neglect an additional linear magnetoelastic coupling involving the antisymmetric strain functions ω_{ij}. This coupling has its origin in the magnetic anisotropy and, consequently, no new coupling constants are required. The importance of these new terms has been clearly demonstrated in the antiferromagnetic phase of MnF_2 by Melcher (1970). He was able to account for differences in the elastic constant c_{44} measured as a function of an applied magnetic field for transverse elastic waves propagating along the [001] and [110] directions. In contrast, the standard theory predicts that the results should be identical in both cases.

1.3 Scope of Thesis

In the present work a rotationally invariant theory of magnetoelastic interactions in ferromagnets is presented. It is shown that ultrasonic measurements of the fractional changes in elastic constant for transverse waves as a function of an applied magnetic field may be used to obtain values for the anisotropy constants as well as the magnetoelastic constants. As in the case of neutron-diffraction and ferromagnetic resonance experiments, a careful analysis of the measurements as a function of the applied field strength and temperature is required.

The concept of rotational invariance is discussed in detail in Chapter 2 and certain important ideas of finite strain theory are outlined. In particular, the finite strain and rotation tensors are defined and the relationship of these tensors to the usual symmetric and antisymmetric strain tensors are derived. In Chapter 3, the methods of constructing a rotationally invariant Hamiltonian in terms of microscopic quantities are outlined. It is also shown how both the static and dynamic effects of magnetoelastic coupling may be investigated using the same Hamiltonian.

Chapter 4 describes the methods employed to obtain the energies of the magnetoelastic normal modes. Both the cases of weak and strong dynamic coupling are considered. In the case of weak coupling, expressions are obtained for

the fractional changes in elastic constant as a function of an applied magnetic field. The expressions for transverse elastic waves, which are either propagating or polarized along the direction of spontaneous magnetization, are written in terms of macroscopic quantities. In this way the values obtained for the various coupling constants may be easily compared with the results of the static measurements such as magnetostriction experiments or magnetic torque experiments.

In Chapter 5 the requirement of rotational invariance is applied to ferromagnetic phases of the rare-earth metals of hcp structure. Expressions are given for the change in elastic constants as function of applied field for impressed sound waves propagating along different crystal axes. The size of the fractional changes $\Delta c/c$ which can be expected are estimated for Gd, Tb, Dy, Ho and Er using available data on magnetocrystalline anisotropy and magnetostriction. Longitudinal sound waves in the paramagnetic region are also discussed.

Chapter 6 applies the same rotationally invariant theory to the heavy rare earth-iron cubic Laves phase compounds RFe_2. The expressions for the fractional change in elastic constant appropriate for cubic symmetry are obtained and estimates for the size of effects that may be expected to be observed in the compounds RFe_2 (R = Tb, Dy, Ho, Er and Tm) are given.

CHAPTER 2

FINITE STRAIN THEORY AND ROTATIONAL INVARIANCE

A correct treatment of magnetoelastic interactions in ordered magnetic crystals must ensure that the total angular momentum (spin plus lattice) of the combined magnetic and elastic systems is conserved. Toupin (1956), Tiersten (1964), Brown (1966) and Melcher (1970) have shown that this requirement is satisfied if the Hamiltonian is constructed using certain invariant quantities. The most convenient ones for a microscopic theory involve the finite strain tensor $E_{\mu\nu}$ and the finite rotation tensor $R_{\mu\nu}$. For this reason some important concepts of finite strain theory are given in the following section (see for example Murnaghan 1951; Truesdell and Toupin 1960).

2.1 Finite Strain Theory

Consider an elastic medium which experiences a deformation. In the initial or undeformed state, the coordinates of a typical particle of the medium with respect to any convenient rectangular cartesian reference frame are denoted by $X_K (K = 1, 2, 3)$. Similarly in the final or

deformed state, the coordinates of the same particle with respect to any other convenient reference frame are denoted by $x_k (k = 1, 2, 3)$. The deformation of a point is thus described by the relation of the coordinates of the same material point in the undeformed and the deformed states. This may be written as

[2.1a] $\underset{\sim}{x} = \underset{\sim}{x}(\underset{\sim}{X})$

or

[2.1b] $\underset{\sim}{X} = \underset{\sim}{X}(\underset{\sim}{x})$.

The X_K are called the material or Lagrangian coordinates and a material description uses these as the independent variables. A spatial description uses the x_k as independent variables and these are referred to as spatial or Eulerian coordinates. The deformation may be described using either method, but a material description will be employed here since the undeformed state has certain known symmetry properties.

The term strain always refers to a change in the relative positions of the material points in a body. If in the initial state we have a particle at (X_1, X_2, X_3) and a neighbouring particle at $(X_1 + dX_1, X_2 + dX_2, X_3 + dX_3)$, then the square of the initial distance between these

particles is

$$[2.2] \quad dS_0^2 = dX_K dX_K$$

where summation over repeated indices is implied here and in the remainder of this chapter. Under the deformation described by eq. [2.1] the particle initially at (X_1, X_2, X_3) moves to (x_1, x_2, x_3) and the square of the final distance to the neighbouring particle is

$$[2.3] \quad dS^2 = dx_k dx_k$$

Since we have chosen the X_K as independent variables, the differentials dx_k are given by the chain rule of differentiation

$$[2.4] \quad dx_k = x_{k,K} dX_K$$

where

$$[2.5] \quad x_{k,K} \equiv \frac{\partial x_k}{\partial X_K}$$

is called the deformation gradient. The finite strain tensor E_{KL} is defined by the relationship of the final distance squared to the initial distance squared as follows

$$[2.6] \quad dS^2 - dS_0^2 \equiv 2E_{KL}dX_K dX_L$$

and may be expressed in terms of the deformation gradients using [2.3] and [2.4]. We find

$$[2.7] \quad E_{KL} = \frac{1}{2}\left[x_{k,K}x_{k,L} - \delta_{K,L}\right] .$$

The finite strain tensor is a measure of strain in the sense that, when $E_{KL} = 0$, the initial and final distances between neighbouring points in the medium are the same. In this case the deformation corresponds to a rigid body displacement or rotation. Note that E_{KL} 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. That is, the finite strain tensor is an invariant with respect to transformations of the spatial coordinates x_k.

Since E_{KL} represents only six independent quantities, whereas the deformation gradients $x_{k,K}$ represent nine independent quantities, the other three degrees of freedom correspond to rotations of the final reference frame with respect to the initial reference frame. These quantities are described by the finite rotation tensor.

The finite rotation tensor is most easily defined in terms of the Green's deformation tensor C_{KL}. This tensor is also a measure of strain and is related to the finite strain tensor as follows

[2.8] $C_{KL} \equiv \delta_{KL} + 2E_{KL} \equiv x_{k,K} x_{k,L}$.

Since C_{KL} is a symmetric tensor, it may be brought to diagonal form at any point in the medium. This means that in both the undeformed and deformed states there exist principal axes of strain. If the final state happens to be the same as the initial state, then C_{KL} is equal to the unit tensor δ_{KL}. In general the final state is not the same and C_{KL} is no longer diagonal. In the principal axes, C_{KL} is a diagonal tensor and we may denote the principal values as C_1, C_2 and C_3. In addition, denoting C as $\bar{C}$ in this set of axes, we may construct a tensor $(\bar{C})^{1/2}$ whose principal values in this same set of axes are $C_1^{1/2}$, $C_2^{1/2}$ and $C_3^{1/2}$ respectively. It is easily verified that $\bar{C} = (\bar{C})^{1/2}(\bar{C})^{1/2}$ and thus in our original set of axes we have

[2.9] $C = U\bar{C}U^{-1} = U(\bar{C})^{1/2} U^{-1} U(\bar{C})^{1/2} U^{-1} = C^{1/2}C^{1/2}$

where U represents the unitary transformation that diagonalizes the tensor C. It is easily verified that $C^{1/2}$ is also a symmetric tensor. The finite rotation tensor R_{Mk} is then defined by the following relation

[2.10] $x_{k,K} = (C^{1/2})_{KM} R_{Mk}$

and it describes the rotation of the principal axes of strain in the initial state to the principal axes in the final state. Hence eq. [2.10] decomposes the deformation into a finite rotation followed by a finite elongation.

This relation may be rewritten as

$$[2.11] \quad R_{Mk} = x_{k,K}(C^{-1/2})_{MK}$$

where $C^{-1/2}$ is a tensor defined in the same manner as $C^{1/2}$. Note that R_{Mk} is not an invariant with respect to the spatial coordinates but transforms as a vector under changes of the final reference frame.

Another quantity of interest is the ratio of the length dS in the deformed state to the length dS_0 in the undeformed state. From eq. [2.6] we have

$$[2.12] \quad dS^2 = (1 + 2E_{KL}\beta_K\beta_L)dS_0^2$$

where β_K are the direction cosines of the line joining two neighbouring particles in the initial state and are given by

$$[2.13] \quad \beta_K = \frac{dX_K}{dS_0} \quad (K = 1, 2, 3) \quad .$$

Taking the square root on both sides of [2.12] we find

$$[2.14] \quad \frac{dS}{dS_0} = [1 + 2E_{KL}\beta_K\beta_L]^{1/2} \quad .$$

In the case where the finite strains can be assumed to be small such that their squares can be neglected, we obtain the following expression

$$[2.15] \quad \frac{dS - dS_0}{dS_0} = E_{KL}\beta_K\beta_L$$

for the change in relative length $\frac{\Delta \ell}{\ell}$ in the direction given by $\beta_K (K = 1, 2, 3)$. This expression may be used to analyze the results of magnetostriction experiments.

In both [2.7] and [2.11] the finite strain tensor and the finite rotation tensor have been expressed in terms of the deformation gradients. In the following section these expressions will be given in terms of the displacement gradients $u_{k,K}$ which are more closely related to the usual infinitesimal measures of strain and rotation.

2.2 Small Displacement Approximation

Under the deformation described by eq. [2.1] the displacement of the particle is defined as

$$[2.16] \quad \underset{\sim}{u} = \underset{\sim}{x} - \underset{\sim}{X} \quad .$$

The displacement gradient $u_{k,K}$ is related to the deformation gradient $x_{k,K}$ as follows

$$[2.17a] \quad u_{k,K} = x_{k,K} - \delta_{k,K}$$

where as in eq. [2.5]

$$[2.17b] \quad u_{k,K} \equiv \frac{\partial u_k}{\partial X_K} \ .$$

In the usual small-strain theory, the displacement gradient is usually decomposed into its symmetric and antisymmetric parts as follows

$$[2.18] \quad u_{k,K} = \varepsilon_{kK} + \omega_{kK}$$

where

$$[2.19a] \quad \varepsilon_{kK} = \varepsilon_{Kk} = \frac{1}{2} [u_{k,K} + u_{K,k}]$$

and

$$[2.19b] \quad \omega_{kK} = -\omega_{Kk} = \frac{1}{2} [u_{k,K} - u_{K,k}]$$

are the infinitesimal strain and rotation tensors respectively.

The finite strain tensor [2.7] may be rewritten using eq. [2.17] as

$$[2.20] \quad E_{KL} = \frac{1}{2}\left[u_{K,L} + u_{L,K}\right] + \frac{1}{2}\, u_{k,K} u_{k,L}$$

$$= \varepsilon_{KL} + \frac{1}{2}\left(\varepsilon_{kK} + \omega_{kK}\right)\left(\varepsilon_{kL} + \omega_{kL}\right) .$$

Thus the infinitesimal strain tensor ε_{KL} is a measure of strain only to first order in the displacement gradients and $\varepsilon_{KL} = 0$ does not necessarily imply $E_{KL} = 0$. A similar expression for the finite rotation tensor may be obtained using [2.11] and [2.17]. In the case of the finite strain tensor the expression obtained was exact. For the finite rotation tensor, the following expansion must be used (Brown 1966)

$$[2.21] \quad \left(C^{-1/2}\right)_{MK} = (1 + 2E)^{-1/2}_{MK} \approx \delta_{MK} - E_{MK} + \frac{3}{2} E_{ML} E_{LK}$$

where terms up to second order in the displacement gradients have been retained. Upon substituting in eq. [2.11] we find

$$[2.22] \quad R_{Mk} = \delta_{Mk} - \omega_{Mk} - \frac{1}{2}\,\omega_{\ell M}\omega_{\ell k} + \frac{1}{2}\,\varepsilon_{\ell M}\omega_{\ell k} - \frac{1}{2}\,\omega_{\ell M}\varepsilon_{\ell k}$$

The infinitesimal rotation tensor ω_{ij} is thus a measure of rotation only to first order in small quantities as would be

expected. It is easily shown that in the linear approximation, where only the first term of [2.20] and the first and second terms of [2.22] are retained, eq. [2.10] corresponds to the usual decomposition of the displacement gradient into its symmetric and antisymmetric parts.

Brown (1966) has shown that within the approximations of linear elasticity theory the distinction between material and spatial coordinates is unnecessary. However, the linear approximations must not be invoked at the beginning, but instead must be applied in a consistent manner which does not violate any of the conservation laws of physics. In the following section we shall discuss the requirement of rotational invariance and also how the results of the customary elasticity theory violate the law of conservation of total angular momentum in a magnetoelastic medium.

2.3 Conservation of Total Angular Momentum

In section 2.1 it was shown that the finite strain tensor E_{KL} is independent of the particular choice of reference frame for the deformed state but that the finite rotation tensor R_{Mk} transforms as a vector under changes in the final state axes. The finite strain tensor can be considered as a scalar product of the vectors $x_{k,K}$ and $x_{k,L}$ in the spatial coordinate system. Another invariant with

respect to the spatial coordinates may be formed by taking the scalar product of the finite rotation tensor and the components S_k ($k = 1, 2, 3$) of spin angular momentum in the deformed state. This quantity may be written as

$$[2.23] \quad S_K^* = R_{Kk} S_k \qquad (K = 1, 2, 3)$$

and is invariant with respect to transformations of the final state axes. Although other invariants such as (Toupin 1956; Tiersten 1964)

$$[2.24] \quad \Pi_K = x_{k,K} S_k \qquad (K = 1, 2, 3)$$

can be formed using the deformation gradients in place of the finite rotation tensor, the S_K^* are more convenient for a microscopic theory since they preserve the spin angular momentum commutation relations (Melcher 1971). This follows from the fact that R_{Kk} is an orthogonal tensor.

If the Hamiltonian which describes the magnetoelastic medium is constructed using the finite strain tensor E_{KL} and the S_K^*, then it will be invariant with respect to arbitrary rotations of the final state coordinate axes, or equivalently of the deformed medium itself. This fact ensures that the total angular momentum (spin plus lattice) of the coupled system is conserved. Following Toupin (1956) the Hamiltonian

is expanded as a power series in these invariants. Note that these invariants do depend on the choice of reference frame in the undeformed state. Thus the point group symmetry of the medium in the undeformed state determines the form of the Hamiltonian. Having ensured complete rotational invariance the usual linear approximations may be applied and the distinction between material and spatial coordinates is no longer necessary. In the case of magnetically anisotropic media, the requirement of rotational invariance leads to the appearance of new terms in the linear magnetoelastic coupling which originate in the magnetic anisotropy and involve the antisymmetric strain functions $\omega_{\mu\nu}$. The standard treatments of magnetoelastic phenomena (Kittel 1958; Akhiezer et al. 1961) do not include these terms because the results of linear elasticity theory are assumed from the beginning. The assumption that the mechanical stress tensor is symmetric leads to magnetoelastic coupling via the symmetric strains $\epsilon_{\mu\nu}$ only. This corresponds to applying the law of conservation of angular momentum separately to the magnetic and elastic systems and violates the conservation of total angular momentum in the coupled system.

CHAPTER 3

STATIC AND DYNAMIC EFFECTS OF MAGNETOELASTIC COUPLING

3.1 Rotationally Invariant Hamiltonian

A rotationally invariant Hamiltonian which describes the coupled magnetic and elastic systems may be constructed using the following invariant quantities (Brown 1966; Melcher 1971)

$$[3.1a] \quad S^*_{j\mu} = R_{\mu\nu} S_{j\nu}$$

$$[3.1b] \quad H^*_{\mu} = R_{\mu\nu} H_{\nu}$$

$$[3.1c] \quad E_{\mu\nu} = \frac{1}{2}\left(\frac{\partial u_\mu}{\partial X_\nu} + \frac{\partial u_\nu}{\partial X_\mu}\right) + \frac{1}{2}\frac{\partial u_\lambda}{\partial X_\mu}\frac{\partial u_\lambda}{\partial X_\nu}$$

where as usual summation over repeated indices is implied and $\underset{\sim}{S}_j$ represents the total electronic angular momentum (spin plus orbital) of the j^{th} atom. The Hamiltonian for the magnetic spin system is taken to consist of Heisenberg exchange, a crystalline electric field of the appropriate symmetry and the Zeeman term. In terms of the invariants defined above we may express it as

$$[3.2]\quad \mathcal{H}_m = -\sum_{i<j} J_{ij}\, \underset{\sim}{S}_i^* \cdot \underset{\sim}{S}_j^* + g u_B \sum_j \underset{\sim}{S}_j^* \cdot \underset{\sim}{H}^* + \sum_j \sum_{\ell,m} B_\ell^m \mathcal{O}_{\ell m}(\underset{\sim}{S}_j^*) \,.$$

The $\mathcal{O}_{\ell m}$ are the spin-operator equivalents tabulated by Buckmaster (1962) and the number of anisotropy constants B_ℓ^m that are required is determined by the point group symmetry of the crystal. The exchange term is assumed to be isotropic since in the limit of long wavelengths the effects of anisotropic exchange may be included with the crystal field.

Following Callen and Callen (1965), the Hamiltonian which describes the coupling between the magnetic and elastic degrees of freedom may be formed by taking products of the symmetry strains and spin functions which transform according to the same irreducible representation of the point group. These spin functions may involve spins on the same atom or spins on different atoms. However, we may include the effects on the coupled modes of such two-ion terms with those of single-ion origin in the limit of long wavelengths. The single-ion magnetoelastic terms may be written as

$$[3.3]\quad \mathcal{H}_{me}^{I} = -\sum_j \sum_\Gamma \sum_{\ell,m} \sum_{s,s'} M_{\ell m}^{\Gamma,s,s'} \sum_r E_r^{\Gamma,s}(j)\, \mathcal{O}_{\ell m}^{\Gamma,r,s'}(\underset{\sim}{S}_j^*)$$

where Γ labels the irreducible representation of order n and r labels the basis functions (r = 1, 2, ..., n). All point groups except those in the cubic system have two sets of basis functions for the totally symmetric representation and

these are labelled by s and s'(s,s' = 1,2). The symmetry strains $E_r^{\Gamma,s}$ are linear combinations of the usual cartesian components $E_{\mu\nu}$ and the number of independent magnetoelastic coupling constants $M_{\ell m}^{\Gamma,s,s'}$ is determined by the particular point group. We shall find that the spin functions $O_{\ell m}^{\Gamma,r,s}$ are most easily expressed in terms of the following combinations of the $O_{\ell m}$:

$$[3.4a] \quad O_{\ell m}^{+} = \frac{1}{2}\,[O_{\ell m} + O_{\ell -m}]$$

$$[3.4b] \quad O_{\ell m}^{-} = \frac{1}{2i}\,[O_{\ell m} - O_{\ell -m}] \quad .$$

Following Chow and Keffer (1973) the strain functions $E_r^{\Gamma,s}(j)$ are to be interpreted as having both a static homogeneous part and a dynamic inhomogeneous part. In this way we may generate both a static and dynamic magnetoelastic coupling from the same Hamiltonian.

The Hamiltonian which describes the elastic system will be taken to have the following form

$$[3.5] \quad \mathcal{H}_e = \frac{1}{2N_a} \sum_j \sum_\Gamma \sum_{s,s'} c_{ss'}^{\Gamma} \sum_r E_r^{\Gamma,s}(j) E_r^{\Gamma,s'}(j)$$

where N_a is the number of atoms per unit volume and the symmetry elastic constants $c_{ss'}^{\Gamma}$ are linear combinations of the usual cartesian elastic constants. As in eq. [3.3] the

$E_r^{\Gamma,s}(j)$ represent the sum of a homogeneous strain portion and an inhomogeneous portion. The homogeneous strains describe the magnetostriction, the effect of homogeneous deformations of the lattice, while the inhomogeneous strains, or phonon modes, correspond to vibrations of the elastic medium about the equilibrium configuration. As will be discussed in section 3.3 these latter strains may be expressed in terms of phonon normal coordinates.

The total magnetoelastic Hamiltonian is given by the following sum

$$[3.6] \quad \mathcal{H} = \mathcal{H}_m + \mathcal{H}_{me}^I + \mathcal{H}_e \ .$$

Since we have ensured complete rotational invariance in the coupled system through the use of the invariants in eq. [3.1], we may now apply the usual approximations of small-strain theory. We shall retain only the first term in eq. [2.20] and the first and second terms in eq. [2.22] when expressing the finite strain and rotation tensors in terms of the usual functions $\varepsilon_{\mu\nu}$ and $\omega_{\mu\nu}$ of small-strain theory. Thus we may rewrite the transformations in eq. [3.1] as follows

$$[3.7a] \quad S_{j\mu}^{*} = S_{j\mu} - \omega_{\mu\nu} S_{j\nu}$$

[3.7b] $H_\mu^* = H_\mu - \omega_{\mu\nu} H_\nu$

[3.7c] $E_{\mu\nu} = \varepsilon_{\mu\nu}$.

When these transformations are substituted into eq. [3.6] we obtain terms similar to those in eqs. [3.2], [3.3] and [3.5] with $\underset{\sim}{S}_j^*$ replaced by $\underset{\sim}{S}_j$ and $E_{\mu\nu}$ replaced by $\varepsilon_{\mu\nu}$ plus new terms involving the antisymmetric rotation tensor $\omega_{\mu\nu}$. These terms originate from the magnetic anisotropy and may be expressed in the following form

[3.8a] $\mathcal{H}_A = \sum_\Gamma \mathcal{H}_A^\Gamma$

[3.8b] $\mathcal{H}_A^\Gamma = \sum_j \sum_{\ell,m} B_\ell^m \sum_r \omega_r^\Gamma(j) O_{\ell m}^{\Gamma,r}(\underset{\sim}{S}_j)$

where Γ labels the irreducible representations according to which the $\omega_{\mu\nu}$ transform and r labels the basis functions of these representations. The actual form of these new linear terms depends upon the point group symmetry of the medium and, in general, the expressions are quite lengthy. In Chapters 5 and 6 we shall give the expressions corresponding to eqs. [3.2], [3.3] and [3.5] for hexagonal-close-packed and cubic symmetry respectively and we shall obtain the additional terms involving the $\omega_{\mu\nu}$. However, a simplified Hamiltonian may be used to illustrate the procedure.

Consider as an example the following Hamiltonian which describes a medium of uniaxial symmetry,

$$[3.9]\quad \mathcal{H} = -\sum_{i<j} J_{ij}\underset{\sim}{S}^{*}_{i}\cdot\underset{\sim}{S}^{*}_{j} + gu_B \sum_j \underset{\sim}{S}^{*}_{j}\cdot\underset{\sim}{H}^{*} + B_2^0 \sum_j \mathcal{O}_{20}(\underset{\sim}{S}^{*}_{j})$$

$$- M_{21}^{\epsilon} \sum_j \{E_{YZ}(j)[i\mathcal{O}_{21}^{+}(\underset{\sim}{S}^{*}_{j})] + E_{XZ}(j)[-i\mathcal{O}_{21}^{-}(\underset{\sim}{S}^{*}_{j})]\}$$

$$+ \frac{c^{\epsilon}}{2N_a} \sum_j \{[E_{YZ}(j)]^2 + [E_{XZ}(j)]^2\} \quad .$$

For simplicity, we have included only the lowest order terms in the spin-operator equivalents and restricted the magnetoelastic and elastic terms to the ϵ-irreducible representation only. The $\mathcal{O}_{\ell m}$ are defined with respect to the crystal axes (X,Y,Z) and the elastic constant c^{ϵ} corresponds to $4c_{44}$ in the usual notation for cubic and hexagonal crystals. In applying the transformations of eq. [3.1], it should be noted that $R_{\mu\nu}$ is an orthogonal tensor and hence both the exchange and Zeeman terms have the same form with respect to either the starred or unstarred quantities. The spin functions $\mathcal{O}_{\ell m}$, however, are anisotropic functions of the spin components and are not identical in terms of the two sets of quantities. For instance, the spin function $\mathcal{O}_{20}(\underset{\sim}{S}_j)$ transforms as follows

$$[3.10]\quad \mathcal{O}_{20}(\underset{\sim}{S}_j^*) = \mathcal{O}_{20}(\underset{\sim}{S}_j) + (6)^{1/2}\{\omega_{YZ}(j)[i\mathcal{O}_{21}^{+}(\underset{\sim}{S}_j)] + \omega_{XZ}(j)[-i\mathcal{O}_{21}^{-}(\underset{\sim}{S}_j)]\}$$

where terms higher than first order in the displacement gradients have been discarded. The spin operators $\mathcal{O}_{21}^{\pm}(\underset{\sim}{S}_j^*)$ also yield additional terms involving the antisymmetric strains $\omega_{\mu\nu}$, but they are multiplied by terms that are already linear in the strains $\varepsilon_{\mu\nu}$ and hence any new linear terms must originate in the magnetic anisotropy terms. Neglecting terms which are quadratic in the infinitesimal strains, we find that the final Hamiltonian has the following form

$$\begin{aligned}[3.11]\quad \mathcal{H} = &- \sum_{i<j} J_{ij}\,\underset{\sim}{S}_i\cdot\underset{\sim}{S}_j + g\mu_B \sum_j \underset{\sim}{S}_j\cdot\underset{\sim}{H} + B_2^0 \sum_j \mathcal{O}_{20}(\underset{\sim}{S}_j)\\ &- M_{21}^{\varepsilon} \sum_j \{\varepsilon_{YZ}(j)[i\mathcal{O}_{21}^{+}(\underset{\sim}{S}_j)] + \varepsilon_{XZ}(j)[-i\mathcal{O}_{21}^{-}(\underset{\sim}{S}_j)]\}\\ &+ \frac{c^{\varepsilon}}{2N_a}\sum_j \{[\varepsilon_{XZ}(j)]^2 + [\varepsilon_{YZ}(j)]^2\}\\ &+ (6)^{1/2} B_2^0 \sum_j \{\omega_{YZ}(j)[i\mathcal{O}_{21}^{+}(\underset{\sim}{S}_j)] + \omega_{XZ}(j)[-i\mathcal{O}_{21}^{-}(\underset{\sim}{S}_j)]\}\,.\end{aligned}$$

These latter terms are not included in the usual treatments of dynamic magnetoelastic coupling in magnetically ordered media (Kittel 1958; Schlömann 1960; Jensen 1971; Vigren and Liu 1972; Chow and Keffer 1973). As we shall discuss in the following sections, these additional terms have no effect upon static equilibrium properties such as magnetostriction, but they are important in the dynamic interaction between magnons and phonons.

3.2 Equilibrium Properties

In the discussion which follows, we shall restrict ourselves to the case of magnetically saturated ferromagnets in which both the magnetic moment distribution and magnetostrictive strains can be assumed to be uniform.

The total magnetoelastic Hamiltonian may be written as the sum of an essentially classical quantity plus terms involving the magnon and phonon operators. This classical quantity describes the total magnetoelastic energy associated with the equilibrium configuration of the magnetic moments and atomic displacements. In the equilibrium state, the directions of the magnetic moments and the values of the static strains due to the magnetic order are determined by minimizing this classical energy with respect to the angles

specifying the magnetization direction and with respect to the homogeneous strain components.

Since we are considering only the homogeneous strains, the three antisymmetric strain functions $\omega_{\mu\nu}$ correspond to homogeneous rotations of the entire medium. Following Callen and Callen (1965) we shall assume that the medium is constrained in such a way that it may not rotate. Hence these homogeneous rotations are zero and only the six symmetric strain components $\varepsilon_{\mu\nu}$, or equivalently, the six symmetry combinations $\varepsilon_r^{\Gamma,s}$ are important.

To illustrate these ideas, consider the simplified Hamiltonian in eq. [3.11] which results from the application of the transformations in eq. [3.1] to eq. [3.9]. We shall write the strain functions as the sum of a homogeneous component and an inhomogeneous component as follows

$$[3.12a]\quad \varepsilon_{\mu\nu}(j) = \varepsilon_{\mu\nu} + \varepsilon_{\mu\nu}(j)$$

$$[3.12b]\quad \omega_{\mu\nu}(j) = \omega_{\mu\nu}(j)$$

where we have assumed that the homogeneous rotations are zero. The energy function which determines the equilibrium properties is obtained by evaluating $\langle \mathcal{H} \rangle$ where the brackets $\langle\ \rangle$ indicate a thermal average with respect to the magnetically saturated state. We have at zero temperature

$$[3.13]\quad \langle \mathcal{H} \rangle = -\frac{N_a S^2}{2} \sum_j J_{ij} - g u_B N_a S\{H_Z \cos\theta + (H_X \cos\phi + H_Y \sin\phi)\sin\theta\} + B_2^0 N_a S(S - \tfrac{1}{2}) P_2^0(\cos\theta) - \frac{\sqrt{6}}{4} M_{21}^{\varepsilon} N_a S(S - \tfrac{1}{2}) \sin 2\theta \{\varepsilon_{YZ} \sin\phi + \varepsilon_{XZ} \cos\phi\} + \frac{1}{2} c^{\varepsilon} \{\varepsilon_{YZ}^2 + \varepsilon_{XZ}^2\}$$

where all terms involving the inhomogeneous strain components vanish in the equilibrium state. The polar angles θ and ϕ specify the direction of magnetization, denoted by z, with respect to the crystal axes and the approximate ground state of the magnetic spins is taken to have $\langle S_j^z \rangle = -S$ at each site.

Carrying out the minimization procedure, we obtain the following equilibrium values for the strain components,

$$[3.14a]\quad \bar{\varepsilon}_{XZ} = \frac{\sqrt{6}}{4} \frac{M_{21}^{\varepsilon}}{c^{\varepsilon}} N_a S(S - \tfrac{1}{2}) \sin 2\theta \cos\phi$$

$$[3.14b]\quad \bar{\varepsilon}_{YZ} = \frac{\sqrt{6}}{4} \frac{M_{21}^{\varepsilon}}{c^{\varepsilon}} N_a S(S - \tfrac{1}{2}) \sin 2\theta \sin\phi .$$

Similarly, the equilibrium values of θ and ϕ are determined from the following conditions

$$[3.15a]\quad \frac{\partial\langle\mathcal{H}\rangle}{\partial\theta} = +gu_B N_a S\{H_Z\sin\theta - (H_X\cos\phi + H_Y\sin\phi)\cos\theta\}$$

$$- 3B_2^0 N_a S(S - \tfrac{1}{2})\sin\theta\cos\theta$$

$$- \frac{\sqrt{6}}{2} M_{21}^{\varepsilon} N_a S(S - \tfrac{1}{2})\cos 2\theta\{\bar{\varepsilon}_{YZ}\sin\phi + \bar{\varepsilon}_{XZ}\cos\phi\}$$

$$= 0$$

$$[3.15b]\quad \frac{\partial\langle\mathcal{H}\rangle}{\partial\phi} = gu_B N_a S\{H_X\sin\phi - H_Y\cos\phi\}\sin\theta$$

$$- \frac{\sqrt{6}}{4} M_{21}^{\varepsilon} N_a S(S - \tfrac{1}{2})\sin 2\theta\{\bar{\varepsilon}_{YZ}\cos\phi - \bar{\varepsilon}_{XZ}\sin\phi\}$$

$$= 0$$

Note that the strains must be considered as independent variables when minimizing with respect to these angles; that is, we do not differentiate the strains in eq. [3.13] with respect to the angles θ and ϕ that occur in [3.14]. The equilibrium configuration is described by the solutions to the above equations.

The strains in eq. [3.14] are functions of the direction cosines of the magnetization and may be used in an expression such as eq. [2.15] to obtain the fractional change in length of the medium in any direction. In this way, values of the various magnetoelastic coupling constants

$M_{\ell m}^{\Gamma,s,s'}$ may be obtained experimentally by analyzing $\frac{\Delta \ell}{\ell}$ both as a function of the direction of magnetization and the direction of observation. The expressions in eq. [3.15] describe the torques exerted on the magnetization components when they depart from the equilibrium direction. These expressions may be used in the analysis of magnetic torque measurements and magnetization measurements to obtain values of the various magnetic anisotropy constants B_ℓ^m (Bozorth 1951). It should be noted that the magnetoelastic constants also enter into the torque equations and make effective contributions to the anisotropy constants. The quantities that are determined in these static measurements are referred to as the anisotropy constants at constant stress. These constants differ from the usual constants B_ℓ^m which are defined at zero strain. Hence in deducing values for the B_ℓ^m, care must be taken to remove the contributions of magnetostriction. This fact may be responsible for the wide scatter of values for the B_ℓ^m in the heavy rare earth metals. As we shall discuss in Appendix A, the contributions of the magnetoelastic terms to the effective anisotropy field are different in the case of dynamic experiments such as ferromagnetic resonance. Thus, before the values obtained for the B_ℓ^m from these various methods may be compared, the effects of magnetoelastic terms must be correctly removed in each case. Brooks (1972) has shown that these effects

are extremely important in the interpretation of the planar anisotropy constant B_6^6 in Tb.

We conclude that the equilibrium properties are not affected by the new linear terms which arise as a consequence of the requirement of rotational invariance. However, when we are considering non-equilibrium properties such as the oscillations about the equilibrium configuration, these new terms are very important. In this case the strains are inhomogeneous and the antisymmetric strains $\omega_{\mu\nu}$ correspond to local rotations of the elastic medium.

3.3 Dynamic Properties

Having determined the equilibrium configuration of the magnetic moments and the atomic displacements, we now consider small oscillations of these quantities about this equilibrium state. A Hamiltonian which describes the dynamic magnetoelastic coupling may be formed by transforming to magnon and phonon operators. The coupled-mode energies may then be obtained using the quantum equations of motion for these operators. This method is equivalent in the limit of long wavelengths to solving the classical equations for the components of magnetization and elastic displacements described in section 1.2.

The spin-wave portion of the Hamiltonian is obtained by standard methods which are equivalent to the transformations of Holstein and Primakoff (1940). However, in addition to the usual magnetic terms contained in $\mathcal{H}_m$, we must also include the terms in $\mathcal{H}_{me}^{I}$ which involve the equilibrium strain components when describing the unperturbed magnon modes. These terms represent the effect of static distortions on the spin-wave excitations. The following Fourier-transformed operators are defined

$$[3.16] \quad S_{\underline{q}}^{\pm} = N_a^{-1/2} \sum_j S_j^{\pm} \exp(\pm i \underline{q} \cdot \underline{r}_j)$$

where the operators $S_j^{\pm} = S_j^x \pm i S_j^y$ represent spin deviations from the equilibrium direction. At zero temperature, the approximate ground state of the magnetic spin system is taken to have $\langle S_j^z \rangle = -S$ for each spin. At finite temperatures, the following expansion of S_j^z is employed (Wortis 1963)

$$[3.17] \quad S_j^z = -S + (2S)^{-1} S_j^+ S_j^- + \cdots$$

In what follows, we shall introduce the effects of temperature through the reduced magnetization, which is defined as

$$[3.18] \quad m = \frac{M(T)}{M(0)} = \frac{\langle \sum_j S_j^z \rangle_T}{\langle \sum_j S_j^z \rangle_{T=0}} \approx 1 - \frac{1}{2N_a S^2} \sum_j \langle S_j^+ S_j^- \rangle \quad .$$

Combining eqs. [3.17] and [3.18] we find that at finite temperatures we may express $\langle S_j^z \rangle$ approximately as follows

$$[3.19] \quad \langle S_j^z \rangle \approx - Sm \quad .$$

Higher powers of S_j^z are treated using the following expression

$$[3.20] \quad (S_j^z)^n \approx (-S)^n + (2S)^{-1} [(1-S)^n - (-S)^n] S_j^+ S_j^-$$

which is obtained from eq. [3.17] using the commutation relations for the spin operators. We shall assume that the operators $S_{\underline{q}}^{\pm}$ obey the following commutation relation

$$[3.21] \quad [S_{\underline{q}_1}^-, S_{\underline{q}_2}^+] = 2Sm \, \delta_{\underline{q}_1 \underline{q}_2}$$

where we have replaced S_j^z by the average value in eq. [3.19].

The unperturbed magnon energies $E_{\underline{q}}$ are now obtained using the equations of motion for the operators $S_{\underline{q}}^+$ and $S_{-\underline{q}}^-$ with the Hamiltonian which includes both $\mathcal{H}_m$ and the terms in $\mathcal{H}_{me}^I$ involving the static strains. The equations have the following form (Brooks 1970; Goodings and Southern 1971)

[3.22a] $[S_{\underline{q}}^{+}, \mathcal{H}] = -A_{\underline{q}} S_{\underline{q}}^{+} - B_{\underline{q}} S_{-\underline{q}}^{-}$

[3.22b] $[S_{-\underline{q}}^{-}, \mathcal{H}] = B_{\underline{q}}^{*} S_{\underline{q}}^{+} + A_{\underline{q}} S_{-\underline{q}}^{-}$.

Hence the correct magnon operators are linear combinations of $S_{\underline{q}}^{+}$ and $S_{-\underline{q}}^{-}$ and may be written as follows

[3.23a] $\alpha_{\underline{q}}^{\dagger} = u_{\underline{q}} S_{\underline{q}}^{+} + v_{\underline{q}} S_{-\underline{q}}^{-}$

[3.23b] $\alpha_{\underline{q}} = u_{\underline{q}}^{*} S_{\underline{q}}^{-} + v_{\underline{q}}^{*} S_{-\underline{q}}^{+}$.

We now impose the condition $[\alpha_{\underline{q}}^{\dagger}, \mathcal{H}] = - E_{\underline{q}} \alpha_{\underline{q}}^{\dagger}$ and we find that the magnon energies are given by

[3.24] $E_{\underline{q}}^{2} = A_{\underline{q}}^{2} - |B_{\underline{q}}|^{2}$.

In addition, the spin operators $S_{\underline{q}}^{+}$ and $S_{-\underline{q}}^{-}$ are related to the magnon operators $\alpha_{\underline{q}}^{\dagger}$ and $\alpha_{\underline{q}}$ as follows

[3.25a] $(S_{\underline{q}}^{+} + S_{-\underline{q}}^{-}) = \left[\frac{2SmE_{\underline{q}}}{A_{\underline{q}} + B_{\underline{q}}}\right]^{1/2} (\alpha_{\underline{q}}^{\dagger} + \alpha_{-\underline{q}})$

[3.25b] $(S_{\underline{q}}^{+} - S_{-\underline{q}}^{-}) = \left[\frac{2SmE_{\underline{q}}}{A_{\underline{q}} - B_{\underline{q}}}\right]^{1/2} (\alpha_{\underline{q}}^{\dagger} - \alpha_{-\underline{q}})$.

The final spin-wave part of the Hamiltonian has the form

$$[3.26]\quad \mathcal{H}_m' = \sum_{\underline{q}} E_{\underline{q}} (\alpha_{\underline{q}}^\dagger \alpha_{\underline{q}} + \tfrac{1}{2})$$

where the magnon operators obey the following commutation relations

$$[\alpha_{\underline{q}_1}, \alpha_{\underline{q}_2}^\dagger] = \delta_{\underline{q}_1 \underline{q}_2}$$

$$[3.27]\quad [\alpha_{\underline{q}_1}, \alpha_{\underline{q}_2}] = 0$$

$$[\alpha_{\underline{q}_1}^\dagger, \alpha_{\underline{q}_2}^\dagger] = 0 \quad .$$

In obtaining the expressions for $A_{\underline{q}}$ and $B_{\underline{q}}$ defined by the equations of motion in eq. [3.22], the following commutation relations for the spin-operator equivalents may be used

$$[3.28a]\quad [S_{-\underline{q}}^-, \sum_j O_{\ell 0}(\underline{S}_j)] = -\frac{\ell(\ell+1)}{2} S(\tfrac{\ell-1}{2}) m^{(\ell-1)(\ell+2)/2} S_{-\underline{q}}^-$$

$$[3.28b]\quad [S_{-\underline{q}}^-, \sum_j O_{\ell 2}(\underline{S}_j)] = \frac{[\ell(\ell+1)(\ell-1)(\ell+2)]^{1/2}}{2} \times S(\tfrac{\ell-1}{2}) m^{(\ell-1)(\ell+2)/2} S_{\underline{q}}^+$$

where

$$[3.29]\quad S(\tfrac{\ell-1}{2}) = (S - \tfrac{1}{2})(S - 1) \ldots (S - \tfrac{\ell-1}{2}) \quad .$$

These operator equivalents are defined with respect to the equilibrium spin direction and hence the Hamiltonian must be transformed to this coordinate system before using the relations above. The following commutators are also non zero, giving constant terms in the equations of motion

$$[3.30] \quad [S^{-}_{-\underset{\sim}{q}}, \sum_{j} O_{\ell 1}(\underset{\sim}{S}_j)] = [\ell(\ell+1)N_a]^{1/2} SS(\tfrac{\ell-1}{2}) m^{\ell(\ell+1)/2} \delta_{\underset{\sim}{q},0} .$$

However, when all these constant terms are grouped together, the magnetic stability conditions, eq. [3.15], cause them to vanish.

The elastic part of the Hamiltonian is obtained by expanding the inhomogeneous strains in terms of the phonon normal coordinates. The unperturbed phonon modes may be expressed as

$$[3.31] \quad \mathcal{H}_e' = \sum_{\underset{\sim}{q}\lambda} \hbar\omega_{\underset{\sim}{q}\lambda} (\beta^{\dagger}_{\underset{\sim}{q}\lambda}\beta_{\underset{\sim}{q}\lambda} + \tfrac{1}{2})$$

where $\beta^{\dagger}_{\underset{\sim}{q}\lambda}$ and $\beta_{\underset{\sim}{q}\lambda}$ are the phonon creation and destruction operators respectively and obey the following commutation relation

$$[3.32] \quad [\beta_{\underset{\sim}{q}_1\lambda_1}, \beta^{\dagger}_{\underset{\sim}{q}_2\lambda_2}] = \delta_{\underset{\sim}{q}_1\underset{\sim}{q}_2}\delta_{\lambda_1\lambda_2} .$$

The unperturbed phonon energy is denoted by $\hbar\omega_{\underline{q}\lambda}$, where $\underline{q}$ refers to the propagation direction and λ refers to the polarization direction. We shall assume that eq. [3.31] describes the acoustic phonon branches only and in this case the elastic constant, or equivalently the sound velocity, is related to the phonon frequency. In the limit of long wavelengths, we have

$$[3.33] \quad \omega_{\underline{q}\lambda}^2 = (c/\rho)|\underline{q}|^2 = v_\lambda^2|\underline{q}|^2$$

where ρ is the density and v_λ is the sound velocity of the phonon branch labelled by λ.

The terms which involve the coupling between the magnons and phonons are obtained from the linear magnetoelastic terms by expanding both the spin functions and the inhomogeneous strain components in terms of the magnon and phonon operators respectively. However, we must include the new linear terms involving the $\omega_{\mu\nu}$ as well as the usual terms contained in $\mathcal{H}_{me}^{I}$. Assuming that the elastic displacements may be expressed as follows (Kittel 1963, Chapter 2)

$$[3.34] \quad \underline{u}(j) = \sum_{\underline{q}\lambda} (2\rho\hbar\omega_{\underline{q}\lambda})^{-1/2} \hat{e}_{\underline{q}\lambda}(\beta_{\underline{q}\lambda} + \beta_{-\underline{q}\lambda}^{\dagger}) e^{i\underline{q}\cdot\underline{r}_j}$$

where $\hat{e}_{\underline{q}\lambda}$ is a unit vector in the polarization direction, we may write the inhomogeneous strain components in the

following form

$$[3.35a]\quad \varepsilon_{\mu\nu}(j) = \frac{i}{2}\sum_{\underline{q}\lambda}(2\rho\hbar\omega_{\underline{q}\lambda})^{-1/2}(\beta_{\underline{q}\lambda}+\beta^{\dagger}_{-\underline{q}\lambda})e^{i\underline{q}\cdot\underline{r}_j} \times [q_\nu\hat{e}^{\mu}_{\underline{q}\lambda}+q_\mu\hat{e}^{\nu}_{\underline{q}\lambda}]$$

$$[3.35b]\quad \omega_{\mu\nu}(j) = \frac{i}{2}\sum_{\underline{q}\lambda}(2\rho\hbar\omega_{\underline{q}\lambda})^{-1/2}(\beta_{\underline{q}\lambda}+\beta^{\dagger}_{-\underline{q}\lambda})e^{i\underline{q}\cdot\underline{r}_j} \times [q_\nu\hat{e}^{\mu}_{\underline{q}\lambda}-q_\mu\hat{e}^{\nu}_{\underline{q}\lambda}] \quad .$$

In the equilibrium magnetic coordinates, the only spin functions which are linear in the magnon operators are $O_{\ell\pm1}(\underline{S}_i)$ and these are linearized as follows

$$[3.36]\quad O_{\ell\pm1}(\underline{S}_j) \tilde{\sim} \pm S(\tfrac{\ell-1}{2})\ \frac{[\ell(\ell+1)]^{1/2}}{2}\ m^{(\ell-1)(\ell+2)/2}\ S^{\pm}_j \quad .$$

In terms of the combinations $O^{\pm}_{\ell1}$ defined in eq. [3.4] we have

$$[3.37a]\quad [iO^{+}_{\ell1}(\underline{S}_j)] \tilde{\sim} -S(\tfrac{\ell-1}{2})\ \frac{[\ell(\ell+1)]^{1/2}}{2}\ m^{(\ell-1)(\ell+2)/2}\ S^{y}_j$$

$$[3.37b]\quad [-iO^{-}_{\ell1}(\underline{S}_j)] \tilde{\sim} -S(\tfrac{\ell-1}{2})\ \frac{[\ell(\ell+1)]^{1/2}}{2}\ m^{(\ell-1)(\ell+2)/2}\ S^{x}_j \quad .$$

When these expressions for the inhomogeneous strain components and the spin functions are substituted into the

linear magnetoelastic coupling terms obtained from $\mathcal{H}_{me}^{I}$ and $\mathcal{H}_{m}$, we find that the magnon-phonon coupling may be written as (Southern and Goodings 1973)

$$[3.38]\quad \mathcal{H}_{mp} = \sum_{\underset{\sim}{q}\lambda} [V_{\underset{\sim}{q}\lambda}\alpha_{\underset{\sim}{q}}(\beta^{\dagger}_{\underset{\sim}{q}\lambda} + \beta_{-\underset{\sim}{q}\lambda}) + V^{*}_{\underset{\sim}{q}\lambda}\alpha^{\dagger}_{\underset{\sim}{q}}(\beta_{\underset{\sim}{q}\lambda} + \beta^{\dagger}_{-\underset{\sim}{q}\lambda})] \quad .$$

We have not included terms of the type considered by Jensen (1971) involving products of two magnon operators and one phonon operator since these should be unimportant at low temperatures. The expressions for the $V_{\underset{\sim}{q}\lambda}$ are complicated functions of both the direction of magnetization and the applied magnetic field.

The total Hamiltonian describing the normal modes of the coupled system is taken as

$$[3.39]\quad \mathcal{H}' = \mathcal{H}_{m}' + \mathcal{H}_{mp} + \mathcal{H}_{e}' \quad .$$

As we shall see in the next chapter, the coupled-mode energies are easily obtained from $\mathcal{H}'$ using an equation of motion technique for the magnon and phonon operators. The important point is that the new linear terms involving the antisymmetric strains $\omega_{\mu\nu}$ are included in $\mathcal{H}_{mp}$. These terms lead to quite surprising predictions for the effect of magnetoelastic interactions on the measured sound velocities of a ferromagnetic material.

CHAPTER 4

MAGNETOELASTIC NORMAL MODES

The Hamiltonian in eq. [3.39] describes the coupled oscillations of the magnetic moments and elastic displacements about their equilibrium configuration. These normal modes of the coupled system may only be described in terms of the unperturbed magnons and phonons when the dynamic interaction is sufficiently weak. If the dynamic interaction is not weak, then these modes possess both magnon and phonon characteristics and should be referred to as magnetoelastic modes. The most striking effects of this interaction are usually thought to occur in the region where the unperturbed magnon and phonon dispersion curves intersect. However, Chow and Keffer (1973) have recently demonstrated that under certain conditions this interaction may also be extremely large in the region of small wavevectors. In this case, the acoustic phonon branch has a strong magnon character and may possibly be excited in an experiment such as magnetic resonance. In the following section we will outline the methods which are used to obtain expressions for the energies of the magnetoelastic modes. We shall also discuss both the case of weak coupling and the case of strong coupling in detail.

4.1 Coupled-Mode Energies

The magnetoelastic-mode energies are easily obtained using an equation of motion technique similar to that employed in section 3.3 to obtain the energies $E_{\underline{q}}$ of the unperturbed magnons. Using the Hamiltonian $\mathcal{H}'$ in eq. [3.39] and the commutation relations in eqs. [3.27] and [3.32], we may write down the equations of motion for the eight operators $\alpha^{\dagger}_{\underline{q}}$, $\alpha_{-\underline{q}}$, $\beta^{\dagger}_{\underline{q}\lambda}$ and $\beta_{-\underline{q}\lambda}$, where λ labels the three acoustic phonon branches. Assuming that the normal mode operators $\gamma^{\dagger}_{\underline{q}\delta}$ and $\gamma_{-\underline{q}\delta}$ (δ = 1, 2, 3, 4) are linear combinations of the eight operators above, we may impose the condition $[\gamma^{\dagger}_{\underline{q}\delta}, \mathcal{H}'] = -\varepsilon_{\underline{q}\delta}\gamma^{\dagger}_{\underline{q}\delta}$ to obtain the coupled mode energies. Since we have one magnon branch and three phonon branches, there are four magnetoelastic modes and the energies are given by the four positive roots of the eight linear equations.

In the general case where all three phonon branches are coupled to the magnon branch with comparable strength, the solution to the eight linear equations is extremely complicated. However, a simple expression may be obtained in the case when only one of the phonon branches is significantly coupled to the magnon mode. In this case two of the magnetoelastic modes are well represented by the two unperturbed phonon branches, while the energies of the remaining magnetoelastic modes are determined by the solutions of four linear equations. The roots of these equations may

be written as

$$[4.1] \quad (\epsilon^{\pm}_{\underline{q}\lambda})^2 = \tfrac{1}{2}(E^2_{\underline{q}} + \hbar^2\omega^2_{\underline{q}\lambda}) \pm \tfrac{1}{2}[(E^2_{\underline{q}} - \hbar^2\omega^2_{\underline{q}\lambda})^2 + 16\hbar\omega_{\underline{q}\lambda}E_{\underline{q}}|V_{\underline{q}\lambda}|^2]^{1/2}$$

where λ labels the particular phonon branch which is coupled to the magnons and the $\pm$ signs refer to the two magnetoelastic modes. In each case, the positive solution is to be taken. Note that the energies of the uniform magnetoelastic modes (i.e., $\underline{q} = 0$) are simply the unperturbed magnon and phonon energies. This follows from the fact that the dynamic interaction vanishes at $\underline{q} = 0$.

In order that we may characterize the strength of the dynamic interaction between the magnons and phonons, we may rewrite eq. [4.1] as follows (Chow and Keffer 1973)

$$[4.2] \quad (\epsilon^{\pm}_{\underline{q}\lambda})^2 = \tfrac{1}{2}(E^2_{\underline{q}} + \hbar^2\omega^2_{\underline{q}\lambda}) \pm \tfrac{1}{2}[(E^2_{\underline{q}} + \hbar^2\omega^2_{\underline{q}\lambda})^2 - 4\hbar^2\omega^2_{\underline{q}\lambda}E'^2_{\underline{q}\lambda}]^{1/2}$$

where the quantity

$$[4.3] \quad (E'_{\underline{q}\lambda}/E_{\underline{q}})^2 = 1 - \frac{4|V_{\underline{q}\lambda}|^2}{E_{\underline{q}}\hbar\omega_{\underline{q}\lambda}}$$

is a measure of the strength of the coupling. When $(E'_{\underline{q}\lambda}/E_{\underline{q}})^2 \approx 1$ the coupling is weak and the two magnetoelastic modes will be similar to the unperturbed modes. At the opposite extreme where $(E'_{\underline{q}\lambda}/E_{\underline{q}})^2 << 1$, the coupling is extremely strong and the modes have both a magnetic and elastic character. These two limits will be considered in the following sections and approximate expressions for the coupled-mode energies will be obtained in each case. In addition, we shall also consider the behaviour of the magnetoelastic modes in the region where the unperturbed modes intersect.

4.2 Weak Coupling Limit

In the case that the coupling may be considered to be weak, we may expand the square root term in eq. [4.1] in terms of small quantities to obtain the following approximate expressions for the coupled-mode energies

$$[4.4] \quad (\epsilon^{\pm}_{\underline{q}\lambda})^2 \approx \frac{1}{2}(E^2_{\underline{q}} + \hbar^2\omega^2_{\underline{q}\lambda}) \pm \frac{1}{2}(E^2_{\underline{q}} - \hbar^2\omega^2_{\underline{q}\lambda}) \pm \frac{4E_{\underline{q}}\hbar\omega_{\underline{q}\lambda}|V_{\underline{q}\lambda}|^2}{(E^2_{\underline{q}} - \hbar^2\omega^2_{\underline{q}\lambda})} .$$

However, this result is only valid in the region far from the point where the unperturbed magnon and phonon branches

intersect. If we restrict our considerations to the case of long wavelength excitations and assume that the energy of the uniform magnon mode E_0 is finite, then we have $\hbar\omega_{\underline{q}\lambda} << E_{\underline{q}}$. In this limit the coupled-mode energies above become

$$[4.5a] \quad (\varepsilon^{+}_{\underline{q}\lambda})^2 \approx E^2_{\underline{q}} + \frac{4\hbar\omega_{\underline{q}\lambda}|V_{\underline{q}\lambda}|^2}{E_{\underline{q}}}$$

$$[4.5b] \quad (\varepsilon^{-}_{\underline{q}\lambda})^2 \approx \hbar^2\omega^2_{\underline{q}\lambda} - \frac{4\hbar\omega_{\underline{q}\lambda}|V_{\underline{q}\lambda}|^2}{E_{\underline{q}}} .$$

The upper mode $\varepsilon^{+}_{\underline{q}\lambda}$ is similar to the unperturbed magnon mode and the lower mode $\varepsilon^{-}_{\underline{q}\lambda}$ is similar to the unperturbed acoustic phonon mode.

If we define an effective elastic constant c^* in the limit of long wavelengths for the lower mode as follows

$$[4.6] \quad (\varepsilon^{-}_{\underline{q}\lambda}/\hbar)^2 = (c^*/\rho)|\underline{q}|^2 ,$$

then we may express eq. [4.5b] as (Southern and Goodings 1973)

$$[4.7] \quad c^* = c - \lim_{\underline{q}\to 0} \frac{4\rho\hbar\omega_{\underline{q}\lambda}|V_{\underline{q}\lambda}|^2}{E_{\underline{q}}|\hbar\underline{q}|^2}$$

where c is the elastic constant for the unperturbed phonon

branch. This expression may be used to analyze measurements of the relative change in elastic constant, or equivalently the sound velocity, as a function of an applied magnetic field. The dependence of the last term in eq. [4.7] on the applied field arises predominantly from the magnon energy $E_{\underline{q}}$. However, both $V_{\underline{q}\lambda}$ and $E_{\underline{q}}$ depend on the direction of magnetization and the applied field direction. In general, these expressions are quite lengthy and complicated.

The expressions for $V_{\underline{q}\lambda}$ and $E_{\underline{q}}$ are simplified, however, when both the direction of magnetization and the applied magnetic field coincide with one another and with one of the crystal symmetry directions. In the case of transverse elastic waves which are either propagating or polarized in the magnetization direction, the latter term in eq. [4.7] may be expressed in terms of macroscopic quantities which are easily derived from a free energy expression for the magnetic spin system. In particular, the uniform magnon-mode energy E_0 may be obtained using the phenomenological macroscopic resonance theory developed by Smit and Belgers (1955) and which is described in Appendix A. For these transverse waves we may express eq. [4.7] in the following form

$$[4.8] \qquad c_T^*(H) = c_T - \frac{[MH_{ME} \pm MH_A]^2}{4M(H_A + H)}$$

where both the direction of magnetization M and the applied field H coincide. The upper sign in eq. [4.8] refers to a transverse wave propagating in the magnetization direction and the lower sign refers to a transverse wave polarized in this direction. Both H_{ME} and H_A depend on the direction of the magnetization and the plane defined by the propagation and polarization directions of the elastic wave. In fact, H_A is the effective anisotropy field experienced by the spin components in this plane when they depart from the easy direction. A general method for obtaining both H_A and H_{ME} from a free energy is outlined in Appendix A and Appendix B respectively. In Chapters 5 and 6 we shall give the appropriate expressions of MH_E and MH_A corresponding to hexagonal-close-packed and cubic symmetry respectively for various directions of the magnetization and for various types of transverse waves.

In order that the effects of domain alignment do not enter the results, we shall assume that some minimum field strength H_0 is required to essentially achieve saturation of the magnetization. Hence the expression which will be used for the analysis of ultrasonic measurements of these elastic constants will have the following form

$$[4.9] \quad c_T^*(H) - c_T^*(H_0) = \frac{M(H - H_0)[H_{ME} \pm H_A]^2}{4(H_A + H)(H_A + H_0)} .$$

It is possible that careful measurements of the various shear wave elastic constants as a function of the applied field strength and temperature may yield values for the various magnetoelastic constants and anisotropy constants that are more reliable than those obtained by other methods.

4.3 Strong Coupling Limit

Before we discuss the case in which $(E'_{\underset{\sim}{q}\lambda}/E_{\underset{\sim}{q}})^2 << 1$, we shall consider first the region where the unperturbed magnon and phonon branches intersect. At the cross-over point, we have $E_{\underset{\sim}{q}} = \hbar\omega_{\underset{\sim}{q}\lambda}$ and the coupled-mode energies in eq. [4.1] become

$$[4.10] \quad (\epsilon^{\pm}_{\underset{\sim}{q}\lambda})^2 = E^2_{\underset{\sim}{q}} \pm 2E_{\underset{\sim}{q}}|V_{\underset{\sim}{q}\lambda}| \ .$$

If we assume that $|V_{\underset{\sim}{q}\lambda}|^2 << E^2_{\underset{\sim}{q}}$, then the energies may be further approximated as follows

$$[4.11] \quad \epsilon^{\pm}_{\underset{\sim}{q}\lambda} = E_{\underset{\sim}{q}} \pm |V_{\underset{\sim}{q}\lambda}|$$

and thus we have gaps of magnitude $2|V_{\underset{\sim}{q}\lambda}|$ appearing in the cross-over region. Both the magnons and phonons are strongly attenuated in this region and the correct normal modes of

the system involve combinations of spin deviations and lattice displacements.

These energy gaps have been observed experimentally in ferromagnetic rare earth metals and alloys (Möller et al. 1967; Nielsen et al. 1970a; Jensen 1971). Expressions for these gaps in terms of the various magnetoelastic coupling constants have also been obtained by Jensen (1971) and Mackintosh and Möller (1972). However, these authors did not include the terms which arise from the requirement of rotational invariance and they obtain the result that the gap expressions are unchanged when the directions of propagation and polarization of transverse phonons are interchanged. The presence of the terms involving the antisymmetric strains $\omega_{\mu\nu}$ leads to different results in these two cases. This difference, as in the case of the elastic constants, may be used to obtain values for the various magnetoelastic and magnetic anisotropy constants.

In general, the following rules may be used to determine which acoustic phonon modes will be coupled to the acoustic magnons and hence to determine which modes will have non-zero gaps. For longitudinal acoustic waves which are propagating parallel or perpendicular to the direction of magnetization, the gaps will be zero. In the case of transverse acoustic waves, the gaps are non-zero only if the direction of propagation or polarization has a component

parallel to the magnetization. These simple rules agree with the conclusions obtained by Mackintosh and Möller (1972) and Nayyar and Sherrington (1972).

In regions far from the cross-over point, we may still have extremely strong coupling between the magnon and phonon branches. In this case we must have $(E'_{\underline{q}\lambda}/E_{\underline{q}})^2 << 1$ and the approximate coupled-mode energies are obtained by expanding eq. [4.2] in terms of small quantities as follows

$$[4.12] \quad (\epsilon^{\pm}_{\underline{q}\lambda})^2 \approx \tfrac{1}{2}(E^2_{\underline{q}} + \hbar^2\omega^2_{\underline{q}\lambda}) \pm \tfrac{1}{2}(E^2_{\underline{q}} + \hbar^2\omega^2_{\underline{q}\lambda}) \mp \frac{\hbar^2\omega^2_{\underline{q}\lambda}E'^2_{\underline{q}\lambda}}{(E^2_{\underline{q}} + \hbar^2\omega^2_{\underline{q}\lambda})} .$$

The normal mode energies are (Chow and Keffer 1973),

$$[4.13a] \quad \epsilon^{+}_{\underline{q}\lambda} \approx (E^2_{\underline{q}} + \hbar^2\omega^2_{\underline{q}\lambda})^{1/2} - \frac{\frac{1}{2}\hbar^2\omega^2_{\underline{q}\lambda}E'^2_{\underline{q}\lambda}}{(E^2_{\underline{q}} + \hbar^2\omega^2_{\underline{q}\lambda})^{3/2}}$$

$$[4.13b] \quad \epsilon^{-}_{\underline{q}\lambda} \approx \hbar\omega_{\underline{q}\lambda} \frac{E'_{\underline{q}\lambda}}{(E^2_{\underline{q}} + \hbar^2\omega^2_{\underline{q}\lambda})^{1/2}} .$$

Chow and Keffer have examined the form of this lower magnetoelastic mode in easy-plane-hexagonal ferromagnets when a magnetic field is applied in the plane along a hard direction. They find that this mode is magnon-like having a

quadratic dependence on wavevector near $\underline{q} = 0$ and they also examine the possibility of exciting these modes by microwaves. The electromagnetic field interacts only with the magnetic spins and thus the absorption due to these modes depends on the amount of magnon character that they possess. The upper magnetoelastic mode remains magnon-like, but in the case of strong coupling, the unperturbed magnon mode does not describe it adequately because the spins and the lattice oscillate together.

In the following chapters we shall investigate the weak coupling case only. We shall apply the results of section 4.2 to the heavy rare earth metals and some of their compounds. The case of strong coupling is extremely complicated and requires further investigation.

CHAPTER 5

HEAVY RARE EARTH METALS

The heavy rare earth metals Tb, Dy, Ho, Er and Tm exhibit large magnetic anisotropy and extremely large magnetostrictive strains at low temperatures and thus the effects of dynamic magnetoelastic coupling are also expected to be appreciable. These metals all have the hexagonal-close-packed (hcp) structure and neutron-diffraction experiments have revealed a rich variety of equilibrium magnetic moment configurations. In each of the various moment arrangements, the moments of the ions lying in a given hexagonal layer are parallel. However, the direction of the moments may change from one layer to the next and this variation of the moment along the hexagonal axis can be described by a wavevector $\underset{\sim}{k}_0$ whose direction is parallel to this axis. Both the crystal field anisotropy and magnetostriction compete with exchange interactions to determine the equilibrium magnetic moment configuration. Exchange favours a spiral spin arrangement while axial anisotropy terms determine whether the arrangement is planar or conical. Even when exchange favours a spiral spin arrangement, planar anisotropy terms and magnetostriction effects can overcome this tendency and give ferromagnetism. A complete

description of the magnetic properties of the rare earth metals is presented in the review article by Cooper (1968b) and in the recent book edited by Elliott (1972). In the results that follow, we shall only consider the ferromagnetic phases of these metals and in this case the spiral wavevector $\underset{\sim}{k}_0$ is zero.

5.1 The Hamiltonian for Hexagonal Symmetry

The magnetic properties of the rare earth metals of hcp structure are well represented by the following Hamiltonian

$$[5.1]\quad \mathcal{H}_m = -\sum_{i<j} J_{ij}\, \underset{\sim}{S}_i^* \cdot \underset{\sim}{S}_j^* + g u_B \sum_j \underset{\sim}{S}_j^* \cdot \underset{\sim}{H}^*$$
$$+ B_2^0 \sum_j O_{20}(\underset{\sim}{S}_j^*) + B_4^0 \sum_j O_{40}(\underset{\sim}{S}_j^*) + B_6^0 \sum_j O_{60}(\underset{\sim}{S}_j^*)$$
$$+ B_6^6 \sum_j [O_{66}(\underset{\sim}{S}_j^*) + O_{6-6}(\underset{\sim}{S}_j^*)] \quad .$$

The first term is the usual Heisenberg exchange coupling predicted by the theory of indirect exchange (see the review article by Kittel 1968). The terms involving the B_L^0 describe a crystal field with axial symmetry while the term in B_6^6 describes the hexagonal anisotropy.

The single-ion magnetoelastic terms have the following form (Goodings and Southern 1971, 1973)

$$[5.2]\quad \mathcal{H}^{I}_{me} = - M^{\alpha,1}_{20} \sum_j E^{\alpha,1}\, O_{20}(\underset{\sim}{S}_j) - M^{\alpha,2}_{20} \sum_j E^{\alpha,2}\, O_{20}(\underset{\sim}{S}_j)$$

$$- M^{\varepsilon}_{21} \sum_j [E^{\varepsilon}_1 (iO^{+}_{21}(\underset{\sim}{S}_j)) + E^{\varepsilon}_2 (-iO^{-}_{21}(\underset{\sim}{S}_j))]$$

$$- M^{\gamma}_{22} \sum_j [E^{\gamma}_1\, O^{+}_{22}(\underset{\sim}{S}_j) + E^{\gamma}_2\, O^{-}_{22}(\underset{\sim}{S}_j)]$$

$$- M^{\gamma}_{44} \sum_j [E^{\gamma}_1\, O^{+}_{44}(\underset{\sim}{S}_j) - E^{\gamma}_2\, O^{-}_{44}(\underset{\sim}{S}_j)]$$

where terms corresponding to $\ell = 6$ as well as certain terms corresponding to $\ell = 4$ have been omitted for the sake of simplicity. In [5.2] the indices α, γ and ε refer to certain combinations of the finite strain components $E_{\mu\nu}$ which transform according to particular irreducible representations of the point group 6/mmm. Choosing a coordinate system $(\xi\eta\zeta)$ coinciding with the a-, b- and c-axes of the crystal, we define

$$[5.3a]\quad E^{\alpha,1} = E_{\xi\xi} + E_{\eta\eta} + E_{\zeta\zeta}$$

$$[5.3b]\quad E^{\alpha,2} = E_{\zeta\zeta} - \tfrac{1}{3} E^{\alpha,1}$$

$$[5.3c]\quad E^{\gamma}_1 = \tfrac{1}{2} (E_{\xi\xi} - E_{\eta\eta})$$

[5.3d] $E_2^{\gamma} = E_{\xi\eta}$

[5.3e] $E_1^{\epsilon} = E_{\eta\zeta}$

[5.3f] $E_2^{\epsilon} = E_{\xi\zeta}$.

The coupling constants in [5.2] are related to those of Callen and Callen (1965) by

$$M_{20}^{\alpha,1} = (3)^{-1/2} B_{12}^{\alpha} \qquad M_{20}^{\alpha,2} = (3)^{-1/2} B_{22}^{\alpha}$$

[5.4]

$$M_{21}^{\epsilon} = (\tfrac{2}{3})^{1/2} B^{\epsilon} \qquad M_{22}^{\gamma} = (\tfrac{2}{3})^{1/2} B^{\gamma} .$$

The effect of two-ion magnetoelastic terms will be described briefly in section 5.3.

The elastic energy associated with the homogeneous strain components has the following form for hexagonal symmetry

$$[5.5] \quad \mathcal{H}_e = \tfrac{1}{2} c_1^{\alpha} (E^{\alpha,1})^2 + c_{12}^{\alpha} E^{\alpha,1} E^{\alpha,2} + \tfrac{1}{2} c_2^{\alpha} (E^{\alpha,2})^2$$

$$+ \tfrac{1}{2} c^{\gamma} [(E_1^{\gamma})^2 + (E_2^{\gamma})^2] + \tfrac{1}{2} c^{\epsilon} [(E_1^{\epsilon})^2 + (E_2^{\epsilon})^2] .$$

The c^{Γ}'s are the elastic stiffness constants which are related to the five independent Cartesian elastic constants by

[5.6a] $c_1^{\alpha} = \frac{1}{9}(2c_{11} + 2c_{12} + 4c_{13} + c_{33})$

[5.6b] $c_2^{\alpha} = \frac{1}{2}(c_{11} + c_{12} - 4c_{13} + 2c_{33})$

[5.6c] $c_{12}^{\alpha} = \frac{1}{3}(-c_{11} - c_{12} + c_{13} + c_{33})$

[5.6d] $c^{\gamma} = 2(c_{11} - c_{12}) = 4c_{66}$

[5.6e] $c^{\epsilon} = 4c_{44}$.

Having ensured complete rotational invariance through the use of the invariants in eq. [3.1], we can now carry out the transformations to the usual spin operators and strain functions of small strain theory. In addition to terms similar to those contained in [5.1], [5.2] and [5.5] with $\underset{\sim}{S}_j$ replaced by $\underset{\sim}{S}_j$ and $E_{\mu\nu}$ replaced by $\epsilon_{\mu\nu}$, we obtain the following terms linear in the $\omega_{\mu\nu}$:

$$\sqrt{6}\, B_2^0 \sum_j [\omega_1^{\epsilon}(iO_{21}^{+}) + \omega_2^{\epsilon}(-iO_{21}^{-})]$$

$$+ 2\sqrt{5}\, B_4^0 \sum_j [\omega_1^{\epsilon}(iO_{41}^{+}) + \omega_2^{\epsilon}(-iO_{41}^{-})]$$

$$+ \sqrt{42}\, B_6^0 \sum_j [\omega_1^{\epsilon}(iO_{61}^{+}) + \omega_2^{\epsilon}(-iO_{61}^{-})]$$

$$+ 2\sqrt{3}\, B_6^6 \sum_j [\omega_1^{\epsilon}(iO_{65}^{+}) + \omega_2^{\epsilon}(+iO_{65}^{-})]$$

$$[5.7] \quad - 12\, B_6^6 \sum_j \omega_2^\gamma \bar{\sigma}_{66} \quad .$$

These extra terms, which have not been included in previous studies of the rare earth metals, involve local rotations of the elastic medium and will be found to have a different effect on shear waves propagating in different directions.

Terms of second order in $\epsilon_{\mu\nu}$ or $\omega_{\mu\nu}$ can also be taken into account without difficulty. For transverse waves propagating along one of the crystal axes and polarized along another crystal axis (for which only one of the derivatives $\partial u_\mu/\partial X_\nu$ or $\partial u_\nu/\partial X_\mu$ is nonzero) these terms contribute to the effective elastic constants. However, these contributions do not depend on the magnetic field in the ferromagnetic phase and consequently do not affect our final results. On the other hand in the paramagnetic region quadratic terms of the type $\epsilon_{\mu\mu}^2$ give rise to field-dependent changes in the elastic constants for longitudinal waves, resulting from the field-induced magnetization in this region. This will be discussed in section 5.5.

5.2 Expressions for Fractional Changes in the Measured Elastic Constants for Hexagonal Symmetry

We now consider the coupling between the acoustic phonons and the magnons, which have two branches in the hcp

structure. This can be derived using the methods outlined in Chapter 3 and the Hamiltonian describing this coupling will have the same form as in eq. [3.38]. The detailed derivation (Jensen 1971) yields no coupling between the acoustic phonons and the higher energy magnon mode, and consequently we shall consider only the lower magnon branch. The expressions for the $V_{q\lambda}$ are very lengthy even for the incomplete Hamiltonian considered here and the full expressions are given in Appendix C.

Allowing for differences in notation our results agree with those given by Nayyar and Sherrington (1972) and Chow and Keffer (1973) except that there are additional terms involving the anisotropy constants which arise from eq. [5.7].

Since our Hamiltonian has the same form as [3.38], the results obtained in the case of weak coupling in section 4.2 may be used to investigate the effects of magnetoelastic interactions on the measured elastic constants of these metals. An important assumption of the results that follow is that the magnetization has been brought into the direction of the applied field, and we denote by H_0 a field large enough both to overcome the effects of magnetocrystalline anisotropy and to give nearly complete alignment of domains. It appears from the recent paper by Palmer and Lee (1972) on

the elastic constants of Dy and Ho that changes in the elastic constants due to domain effects can be of the order of 1% and consequently can mask the "intrinsic" field dependence with which we are concerned. Another instance is the dip in c_{33} as a function of temperature measured by Long, Wazzan and Stern (1969) in Gd near 220°K. This has been interpreted by Levinson and Shtrikman (1971) as essentially arising from the alignment of domains below about 5 kOe. Thus it is important that H_0 be large enough to achieve complete saturation of the magnetization in the direction of the applied field. The minimum value of H_0 will also depend on the demagnetizing fields in the sample. For example, in a Gd crystal of rectangular cross section Moran and Lüthi (1970) found that the magnetization did not saturate until H = 10 kOe.

Below we give the results for the changes in elastic constants as a function of magnetic field based on eq. [4.9] of Chapter 4. The case in which the field is applied along the c-axis and the case in which it is applied along either an a- or b-axis will be considered separately. It is convenient to introduce the following definitions, with N_a the number of atoms per unit volume:

[5.8a] $b_2 = N_a B_2^0 SS(\frac{1}{2})$

[5.8b] $b_4 = N_a B_4^0 SS(\frac{3}{2})$

[5.8c] $b_6 = N_a B_6^0 SS(\frac{5}{2})$

[5.8d] $b_6^6 = N_a[(231)^{1/2}/16]B_6^6 SS(\frac{5}{2})$

[5.8e] $b_2^\epsilon = N_a(\frac{3}{2})^{1/2} M_{21}^\epsilon SS(\frac{1}{2}) = \frac{1}{2} c^\epsilon H(0)$

[5.8f] $b_2^\gamma = N_a(\frac{3}{2})^{1/2} M_{22}^\gamma SS(\frac{1}{2}) = 2c^\gamma C(0)$

[5.8g] $b_4^\gamma = N_a(\frac{70}{16})^{1/2} M_{44}^\gamma SS(\frac{3}{2}) = -2c^\gamma A(0)$

[5.8h] $S(n) = (S - \frac{1}{2})(S - 1) \ldots (S - n)$

[5.8i] $h = N_a g u_B HS = M(0)H$

[5.8j] $h_0 = N_a g u_B H_0 S = M(0)H_0$.

The constants H(0), C(0) and A(0) occuring in [5.8e] - [5.8g] are the magnetostriction constants of Mason (1954) at T = 0.

For the case in which H is applied parallel to the c-axis of the sample, we find that c_{11}, c_{33} and c_{66} are not affected by the applied field in the ferromagnetic region. For the field dependence of c_{44} we find,

[5.9a] $$c_{44}^*(H) - c_{44}^*(H_0) = \frac{m(h - h_0)(H_{ME} \pm H_A)^2}{4(H_A + H)(H_A + H_0)}$$

where

$$[5.9b] \quad M(T)H_A = -3b_2 m^3 - 10b_4 m^{10} - 21b_6 m^{21}$$

$$[5.9c] \quad M(T)H_{ME} = b_2^{\varepsilon} m^3 = \frac{c^{\varepsilon} H(T)}{2} \quad .$$

Here $m = M(T)/M(0)$ is the reduced magnetization. The upper sign in [5.9a] is the result for a wave propagating along the c-axis and polarized along either the a- or b-directions, while the lower sign is for propagation along either the a-axis or b-axis and polarized in the c-direction. The usual small-strain theory yields the same result for both types of wave, the last factor in the numerator of [5.9a] being simply H_{ME}^2.

For H parallel to the a-axis or b-axis, c_{11} and c_{33} are not affected by the applied field in the ferromagnetic region. The behaviour of c_{44} is again given by

$$[5.10a] \quad c_{44}^*(H) - c_{44}^*(H_0) = \frac{m(h - h_0)(H_{ME} \pm H_A)^2}{4(H_A + H)(H_A + H_0)}$$

where

$$[5.10b] \quad MH_A = 3b_2 m^3 - (\tfrac{15}{2}) b_4 m^{10} + (\tfrac{105}{8}) b_6 m^{21} - 6b_6^6 \cos 6\phi\, m^{21}$$
$$+ [2(b_2^{\gamma} m^3)^2 + (b_4^{\gamma} m^{10})^2 + 3(b_2^{\gamma} m^3)(b_4^{\gamma} m^{10}) \cos 6\phi]/4c^{\gamma}$$

continued...

$$= 3b_2m^3 - (\tfrac{15}{2})b_4m^{10} + (\tfrac{105}{8})b_6m^{21} - 6b_6^6\cos 6\phi m^{21}$$

$$+ c^{\gamma}[2C^2 + A^2 - 3AC\cos 6\phi]$$

[5.10c] $$MH_{ME} = - b_2^{\varepsilon}m^3 = - \frac{c^{\varepsilon}H(T)}{2} \quad .$$

Here ϕ is zero for H//a and $\pi/2$ for H//b. The upper sign in [5.10a] is the result for a sound wave propagating along the direction of $\underline{H}$ and polarized along the c-axis. The lower sign is for propagation along the c-axis and polarized along the direction of $\underline{H}$. Again the small strain theory gives the same result for both types of wave, the last factor in the numerator of [5.10a] being simply H_{ME}^2.

For transverse waves propagating in the a- or b-directions and polarized in the basal plane, we find the result,

[5.11a] $$c_{66}^*(H) - c_{66}^*(H_0) = \frac{m(h - h_0)(H_{ME} \pm H_A)^2}{4(H_A + H)(H_A + H_0)}$$

where

[5.11b] $$MH_A = - 36b_6^6\cos 6\phi m^{21} + [4(b_2^{\gamma}m^3)^2 + 4(b_2^{\gamma}m^{10})^2$$

$$+ 10(b_2^{\gamma}m^3)(b_4^{\gamma}m^{10})\cos 6\phi]/4c^{\gamma}$$

$$= - 36b_6^6\cos 6\phi m^{21} + c^{\gamma}[4C^2 + 4A^2 - 10AC\cos 6\phi]$$

$$[5.11c]\quad MH_{ME} = + b_2^{\gamma} m^3 \cos 2\phi - b_4^{\gamma} m^{10} \cos 4\phi$$

$$= + 2c^{\gamma} [C\cos 2\phi + A\cos 4\phi] \quad .$$

The upper sign in [5.11a] is the result for propagation along $\underset{\sim}{H}$ and the lower sign is for polarization along $\underset{\sim}{H}$. ϕ is zero for H//a and $\pi/2$ for H//b. Small strain theory results in the same expression for both types of wave, the last factor in the numerator of [5.11a] being simply $(H_{ME})^2$ for either orientation of the applied field.

Although these results were obtained from the Hamiltonian in the form of eq. [3.38], we have verified that the same results are obtained by solving the macroscopic equations [1.10] and [1.5] for the transverse components of magnetization and the elastic displacements respectively, having first rewritten the Hamiltonian of eqs. [5.1] and [5.2] together with the elastic energy in terms of macroscopic quantities. In the case in which the spins are aligned along the c-axis, the calculation for the present two-sublattice ferromagnet is only slightly different from that carried out by Melcher (1971) for the two-sublattice uniaxial antiferromagnet.

5.3 The Effect of Two-Ion Magnetoelastic Interactions

In addition to the one-ion magnetoelastic terms of eq. [5.2] there occur two-ion magnetoelastic terms which come primarily from the strain dependence of the exchange interactions. In this section we examine briefly how the inclusion of these terms affects the results of the preceding section.

To second order in the angular momentum invariants $S^*_{i\mu}$ and first order in the finite strains $E_{\mu\nu}$ the two-ion terms are (Callen and Callen 1965; Goodings and Southern 1971)

$$[5.12a]\quad \mathcal{H}^{II}_{me} = \sum_{i<j} \mathcal{H}^{II}_{me}(i,j)$$

$$[5.12b]\quad \mathcal{H}^{II}_{me}(i,j) = - D^{\alpha}_{11ij}\, E^{\alpha,1}\, \underset{\sim}{S}^*_i \cdot \underset{\sim}{S}^*_j - D^{\alpha}_{12ij}\, E^{\alpha,1}(\sqrt{3}/2)$$

$$\times (S^*_{i\zeta} S^*_{j\zeta} - \tfrac{1}{3}\, \underset{\sim}{S}^*_i \cdot \underset{\sim}{S}^*_j) - D^{\alpha}_{21ij}\, E^{\alpha,2}\, \underset{\sim}{S}^*_i \cdot \underset{\sim}{S}^*_j$$

$$- D^{\alpha}_{22ij}\, E^{\alpha,2}(\sqrt{3}/2)(S^*_{i\zeta} S^*_{j\zeta} - \tfrac{1}{3}\, \underset{\sim}{S}^*_i \cdot \underset{\sim}{S}^*_j)$$

$$- D^{\gamma}_{ij}[E^{\gamma}_1 \tfrac{1}{2}(S^*_{i\xi} S^*_{j\xi} - S^*_{i\eta} S^*_{j\eta})$$

$$+ E^{\gamma}_2 \tfrac{1}{2}(S^*_{i\xi} S^*_{j\eta} + S^*_{i\eta} S^*_{j\xi})] - D^{\epsilon}_{ij}[E^{\epsilon}_1 \tfrac{1}{2}(S^*_{i\eta} S^*_{j\zeta}$$

$$+ S^*_{i\zeta} S_{j\eta}) + E^{\epsilon}_2 \tfrac{1}{2}(S^*_{i\xi} S^*_{j\zeta} + S^*_{i\zeta} S^*_{j\xi})] \cdot$$

The main way in which these terms affect the results of the preceding section is to cause the following replacements wherever they occur in eqs. [5.9] - [5.11]:

$$[5.13a]\quad b_2^{\gamma} m^3 \rightarrow b_2^{\gamma} m^3 + d^{\gamma} m^2$$

$$[5.13b]\quad b_2^{\varepsilon} m^3 \rightarrow b_2^{\varepsilon} m^3 + d^{\varepsilon} m^2$$

where

$$[5.13c]\quad d^{\gamma} = \frac{1}{2} N_a S^2 \sum_j D_{ij}^{\gamma}$$

$$[5.13d]\quad d^{\varepsilon} = \frac{1}{2} N_a S^2 \sum_j D_{ij}^{\varepsilon} \,.$$

In the latter definitions the sum over j is over all neighbours of i, not just those on one sublattice.

In practice it will be difficult to separate these one-ion and two-ion magnetoelastic contributions since their temperature dependence is quite similar. In the calculations to be described in the next section we have regarded the two-ion terms as making effective contributions to $b_2^{\gamma} m^3$ and $b_2^{\varepsilon} m^3$, the size of which is unknown. However, since the strain dependence of the Heisenberg exchange terms leads only to terms of α-symmetry in [5.12], the two-ion terms in d^{γ} and d^{ε} must have their origin in some higher-order coupling

between the angular momenta $\underset{\sim}{S}_i$ and $\underset{\sim}{S}_j$, and thus there is reason to believe that they will be much smaller than b_2^γ and b_2^ε.

5.4 Estimates for Gd, Tb, Dy, Ho and Er

Measurements of the changes in elastic constants as a function of the applied field can be analyzed using the expressions of section 5.2 in order to obtain accurate values of the various magnetoelastic constants and, in favourable cases, of the anisotropy constants as well. In this section we turn this process around and use the available data for the magnetoelastic constants and the anisotropy constants to estimate the size of the effects which can be expected to be measured experimentally. The discussion is restricted, of course, to temperature ranges in which ferromagnetism occurs, the temperature dependence having been expressed through powers of the reduced magnetization in the last two sections. In fact in making the estimates that follow we have replaced $m^{\ell(\ell+1)/2}$ by the (normalized) hyperbolic Bessel function $I_{\ell+1/2}(\lambda^{-1}(m))$ in order to improve the results in the region of higher temperatures (Callen and Callen 1966). Throughout this section we shall be concerned with transverse acoustic waves only. Longitudinal waves will be treated in the following section.

Estimates of the various anisotropy constants obtained from torque measurements and magnetization curves are given in Table 1 for Gd, Tb, Dy, Ho and Er. The values used in the calculations of Figs. 1 - 7 were those determined from magnetization curves by Peron, Hug and Pauthenet (1970). Tables 2 and 3 give estimates of b_2^ϵ, b_2^γ and b_4^γ based on the available magnetostrictive data.

Gadolinium

Fig. 1 shows the estimated fractional change in the elastic constant c_{44} for a magnetic field along the a-axis. The ordinate is $[c_{44}^*(H) - c_{44}^*(H_0)]/c_{44}^*$ calculated from eq. [5.10], it being immaterial what strength of H is used for c_{44}^* in the denominator. A value of H_0 = 10 kOe was chosen, which is probably large enough to saturate the magnetization and sufficient to mask the undesirable effect of nonuniform demagnetizing fields in nonellipsoidal samples. The solid curves are the results for transverse waves propagating along the c-axis and polarized in the a-direction while the dashed curves are for waves propagating along the a-axis and polarized in the c-direction. Our calculations show that as m decreases from 1.0 to 0.3, $\Delta c_{44}/c_{44}$ decreases by about two orders of magnitude. Thus the most accurate values of b_2^ϵ will be obtained from measurements at low temperatures. A detailed study of the temperature dependence of the results

TABLE 1

ESTIMATES OF THE ANISOTROPY CONSTANTS b_2, b_4, b_6, b_6^6 AND THE MAGNETIC FIELD PARAMETER h CORRESPONDING TO H = 10 kOe

(10^6 ergs/cm^3)

	N_a (10^{22} atoms/cm^3)	b_2	b_4	b_6	b_6^6	h	References
Gd	3.04	-1.3	+0.69		-.0064	19.7	Feron et al. (1970), Graham (1966)
	3.04	-0.55	+0.64	-.061	-.0064	19.7	Brooks and Goodings (1968), Graham (1967)
Tb	3.15	+565	+46		+1.85	26.3	Feron et al. (1970)
	3.15	+550			+2.42	26.3	Rhyne and Clark (1967)
	3.15	+450				26.3	Rhyne et al. (1968)
Dy	3.17	+550	-54		-11	29.4	Feron et al. (1970)
	3.17	+550			-7.6	29.4	Rhyne and Clark (1967)
	3.17	+490				29.4	Rhyne et al. (1968)
Ho	3.21	+416	+177		+27	29.8	Feron et al. (1970)
	3.21	+ 63	+48		+.21	29.8	Bozorth et al. (1968)
Er	3.26	-180[a]	-27[a]	+62[a]	-38[a]	27.2	Bozorth et al. (1972)

[a] Deduced from analysis of HoEr alloys.

Fig. 1: $\Delta c_{44}/c_{44}$ as a function of $(H - H_0)/H_0$ for Gd calculated from eq. [5.10a] for a magnetic field along the a-axis. H_0 = 10 kOe, $b_2^{\epsilon} = 0.54 \times 10^8$ ergs/cm^3 and $c_{44} = 0.226 \times 10^{12}$ ergs/cm^3. Solid curves are for propagation along the c-axis and dashed curves are for propagation along the a-axis. Curves are shown for values of the reduced magnetization m ranging from 0.85 to 0.5.

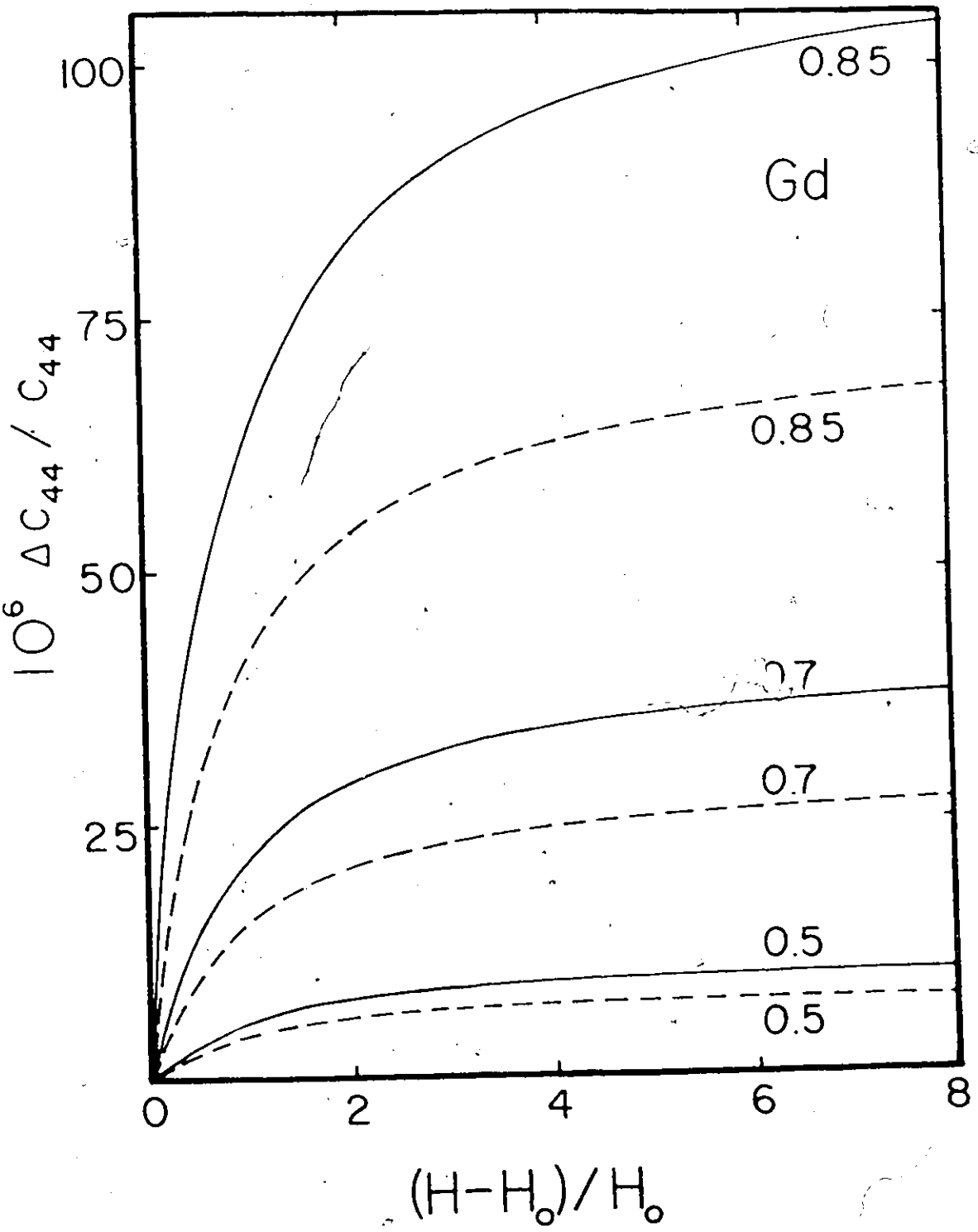
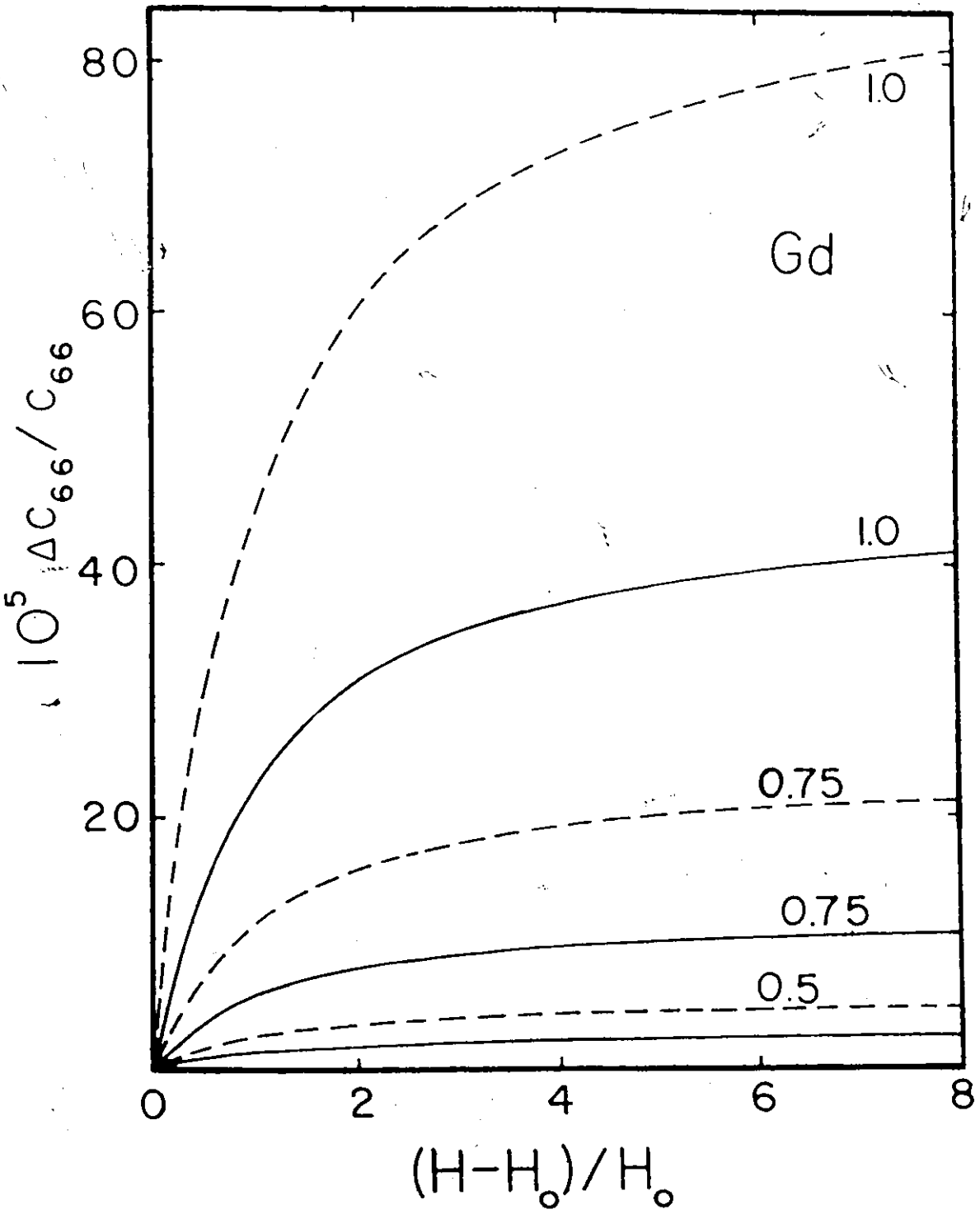

100
75
50
25
0
2
4
6
8
0.85
Gd
0.85
0.7
0.7
0.5
0.5
$10^6\ \Delta C_{44} / C_{44}$
$(H-H_o)/H_o$

Fig. 2: $\Delta c_{66}/c_{66}$ as a function of $(H - H_0)/H_0$ for Gd calculated from eq. [5.11a] for a magnetic field along the a-axis. $b_2^\gamma = 0.92 \times 10^8$ ergs/cm^3 and $c_{66} = 0.229 \times 10^{12}$ ergs/cm^3. The dashed curves are for $H_0 = 5$ kOe while the solid curves are for $H_0 = 10$ kOe. Curves are shown for values of m ranging from 1.0 to 0.5.


80
60
40
20
0
2
4
6
8
1.0
Gd
1.0
0.75
0.75
0.5
$10^5 \Delta C_{66} / C_{66}$
$(H-H_o)/H_o$

might also yield values of the anisotropy constants b_2 and b_4 contained in the effective anisotropy field H_A.

Estimates of the fractional change in c_{66} based on eq. [5.11] for a field H along the a-axis are shown in Fig. 2. The results for transverse waves propagating along the a- or b-axes and polarized in the plane are indistinguishable from each other, as a consequence of the weak hexagonal anisotropy. To show the effect of choosing different values for the minimum field strength H_0, curves have been plotted for H_0 = 5 kOe and H_0 = 10kOe with m ranging from 1.0 down to 0.5. Again the greatest fractional change is expected at low temperatures.

It is worth noting that measurements of $\Delta c_{66}/c_{66}$ for H//b when compared with the results for H//a provide the possibility of obtaining independent values for b_2^γ and b_4^γ, as may be seen from eq. [5.11c] setting $\phi = \pi/2$ and $\phi = 0$ in the two cases.

Terbium and Dysprosium

As the anisotropy constants and magnetoelastic constants for Tb and Dy are about two orders of magnitude larger than for Gd, the fractional changes in elastic constants $\Delta c/c$ are of the order 10^{-3} to 10^{-2} compared with about 10^{-5} to 10^{-4} in the case of Gd (Moran and Lüthi 1970). Figs. 3 and 4 show calculations of $\Delta c_{44}/c_{44}$ for Tb and Dy

TABLE 2

ESTIMATES OF b_2^ε

	c_{44} (10^{12} ergs/cm^3)	Temp. of measurement	Ref.	$\lambda^\varepsilon(0) = \frac{H(0)_{Mason}}{2}$	Ref.	$b_2^\varepsilon = 4c_{44}\lambda^\varepsilon(0)$ (10^8 ergs/cm^3)
Gd	.238	4 K	1	$.04 \times 10^{-3}$	5	.57
	.213	300 K	2	$.04 \times 10^{-3}$	5	.51
Tb	.228	300 K	2	20×10^{-3}	6	180
Dy	.270	0 K	3	9.16×10^{-3}	7	59.4
	.243	298 K	4	9.16×10^{-3}	7	53.5

1 Fisher and Dever (1968)
2 Pollina and Lüthi (1969)
3 Rosen and Klimker (1970)
4 Fisher and Dever (1967)
5 Alstad and Legvold (1964)
6 DuPlessis (1968)
7 Clark et al. (1965)

Fig. 3: $\Delta c_{44}/c_{44}$ as a function of $(H - H_0)/H_0$ for Tb calculated from eq. [5.10a] for a magnetic field along the b-axis. H_0 = 10 kOe, $b_2^{\epsilon} = 180 \times 10^8$ ergs/cm^3 and $c_{44} = 0.228 \times 10^{12}$ ergs/cm^3. Solid curves are for propagation along the c-axis and dashed curves are for propagation along the direction of the magnetic field. Curves are shown for values of m ranging from 1.0 to 0.3.

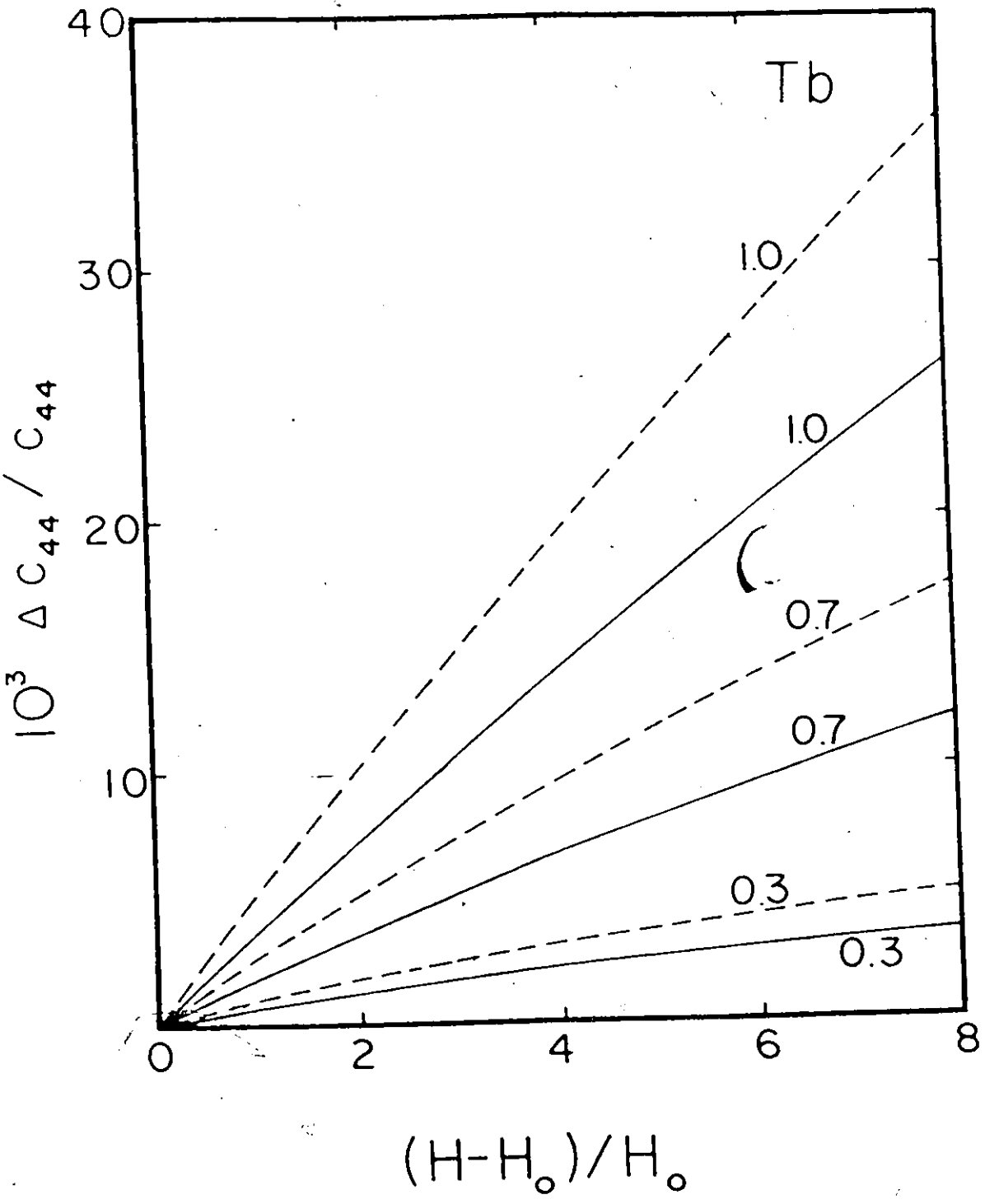
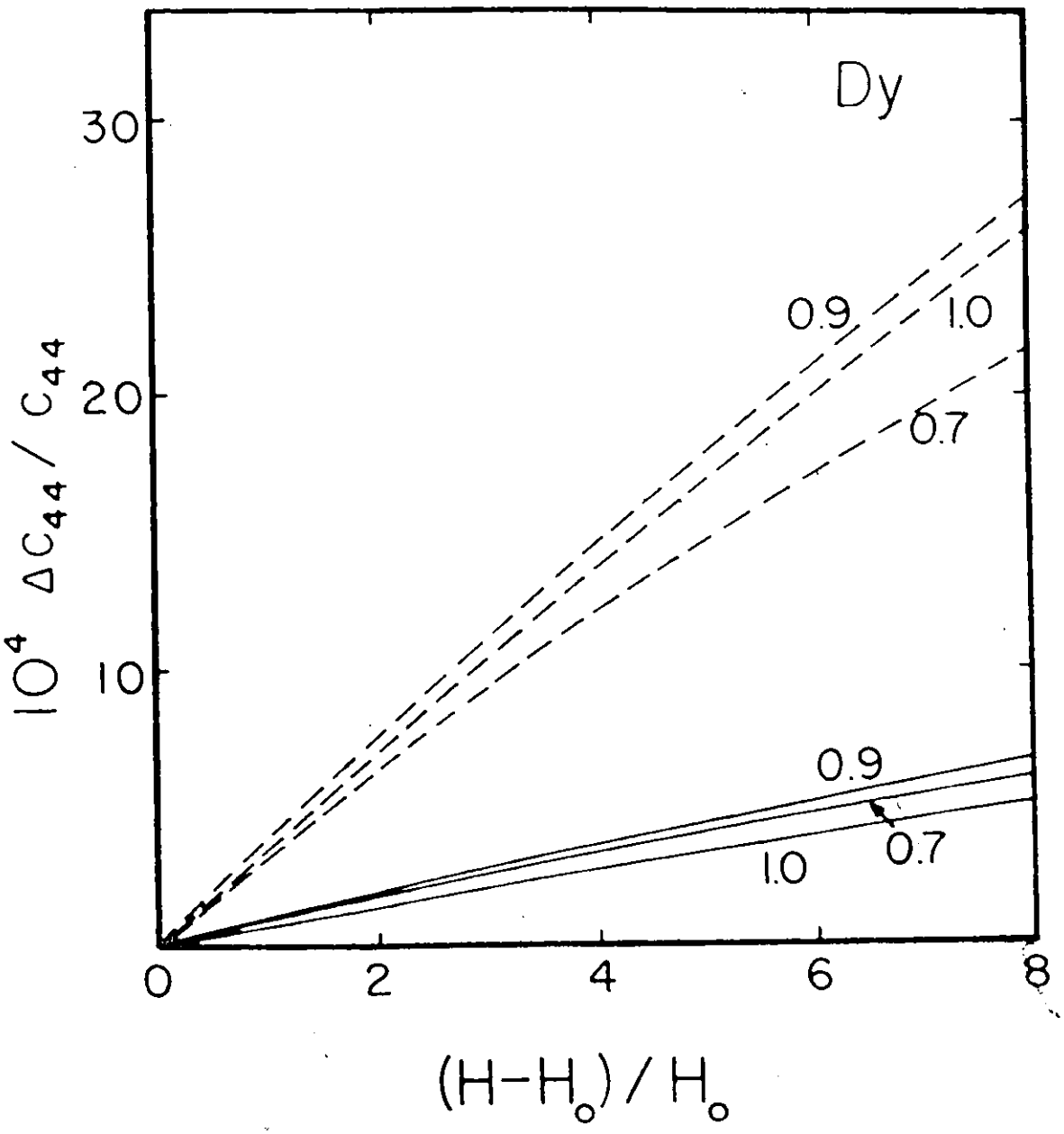

Tb
1.0
1.0
0.7
0.7
0.3
0.3
40
30
20
10
0
2
4
6
8
$10^3 \Delta C_{44} / C_{44}$
$(H-H_o)/H_o$

Fig. 4: $\Delta c_{44}/c_{44}$ as a function of $(H - H_0)/H_0$ for Dy calculated from eq. [5.10a] for a magnetic field along the a-axis. H_0 = 10 kOe, $b_2^{\epsilon} = 56 \times 10^8$ ergs/cm^3 and $c_{44} = 0.257 \times 10^{12}$ ergs/cm^3. Solid curves are for propagation along the c-axis and dashed curves are for propagation along the direction of the magnetic field. Curves are shown for values of m ranging from 1.0 to 0.7.


Dy
30
20
10
0
2
4
6
8
0.9
1.0
0.7
0.9
1.0
0.7
$10^4\ \Delta C_{44} / C_{44}$
$(H-H_o)/H_o$

respectively based on eq. [5.10]. The magnetic field was taken to be along the easy direction (b-axis for Tb, a-axis for Dy) and H_0 was chosen to be 10 kOe in each case. The solid curves are for waves propagating along the c-axis and polarized in the easy direction, while the dashed curves are for waves propagating along the easy direction and polarized in the c-direction. In comparison with Gd, these curves are much more nearly linear because the anisotropy terms dominate the effect of the field in the denominator of eq. [5.10a]. For Tb the largest fractional changes occur at low temperatures while for Dy the largest changes occur in the region of m = 0.85 as a result of a competition among the various factors in the numerator and denominator of eq. [5.10a].

In Figs. 5 and 6 we have plotted estimates of $\Delta c_{66}/c_{66}$ for Tb and Dy respectively based on eq. [5.11]. The magnetic field was taken to be along the easy direction in each case and H_0 was chosen to be 10 kOe. The solid curves are for polarization along the direction of $\underset{\sim}{H}$ while the dashed curves are for propagation in this direction. The small difference between these two sets of curves results from the fact that the hexagonal anisotropy is at least 20 times smaller than H_{ME} near T = 0. As H_A falls to zero with increasing temperature the two sets of curves become indistinguishable. It can be seen from Figs. 5 and 6 that the maximum changes in c_{66} occur around m = 0.75.

TABLE 3

ESTIMATES OF b_2^γ AND b_4^γ

(10^8 ergs/cm^3)

	c_{66} (10^{12} ergs/cm^3)	Temp. of measurement	Ref.	$\lambda^{\gamma,2}(0)$ = $2C(0)_{Mason}$	$\lambda^{\gamma,4}(0)$ = $-2A(0)_{Mason}$	Ref.	b_2^γ = $4c_{66}\lambda^{\gamma,2}(0)$	b_4^γ = $4c_{66}\lambda^{\gamma,4}(0)$
Gd	.229	4 K	1	$.10 \times 10^{-3}$		6	.92	
Tb	.22	-	2	8.7×10^{-3}	-4.3×10^{-3}	7	77	-38
	.22	-		8.5×10^{-3}	-5.0×10^{-3}	8	76	-44
Dy	.283	0 K	3	8.5×10^{-3}		9	96	
	.243	298 K	4	8.5×10^{-3}		9	82	
Ho	.308[a]	4 K	5	2.5×10^{-3}		10	31	
Er	.303	63 K	4	-5.4×10^{-3}		11	-65	
	.279	298 K	4	-5.4×10^{-3}		11	-60	

[a] shear modulus measured in a polycrystalline sample

1 Fisher and Dever (1968)
2 Cooper (1968a)
3 Rosen and Klimker (1970)
4 Fisher and Dever (1967)
5 Rosen (1967)
6 Alstad and Legvold (1964)
7 Rhyne and Legvold (1965a)
8 DuPlessis (1968)
9 Clark et al. (1965)
10 Rhyne et al. (1967)
11 Rhyne and Legvold (1965b)

Fig. 5: $\Delta c_{66}/c_{66}$ as a function of $(H - H_0)/H_0$ for Tb calculated from eq. [5.11a] for a magnetic field along the b-axis. $H_0 = 10$ kOe, $b_2^\gamma = 77 \times 10^8$ ergs/cm^3, $b_4^\gamma = -41 \times 10^8$ ergs/cm^3 and $c_{66} = 0.22 \times 10^{12}$ ergs/cm^3. Solid curves are for propagation along the a-axis and dashed curves are for propagation along the b-axis. Curves are shown for values of m ranging from 1.0 to 0.5.

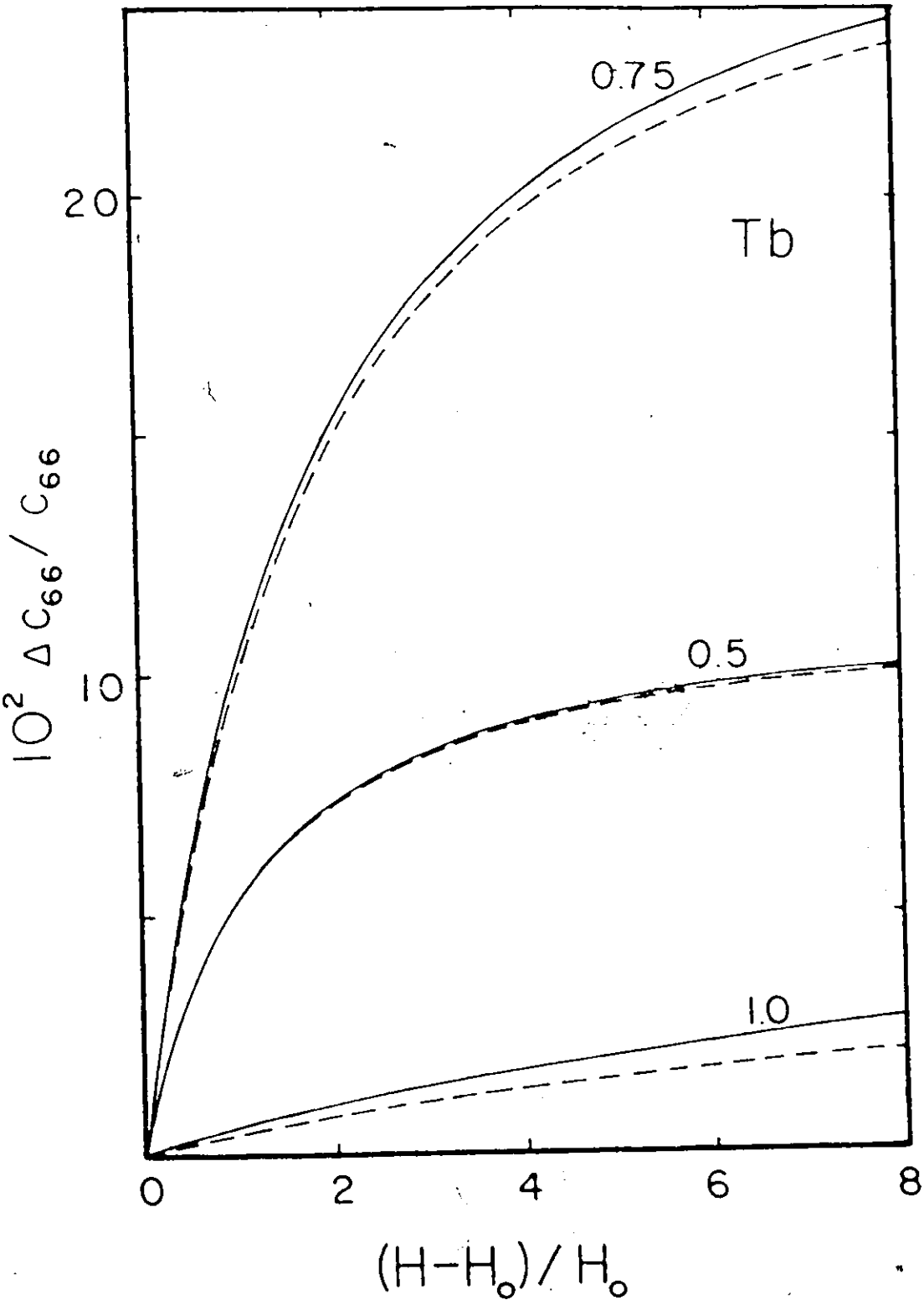
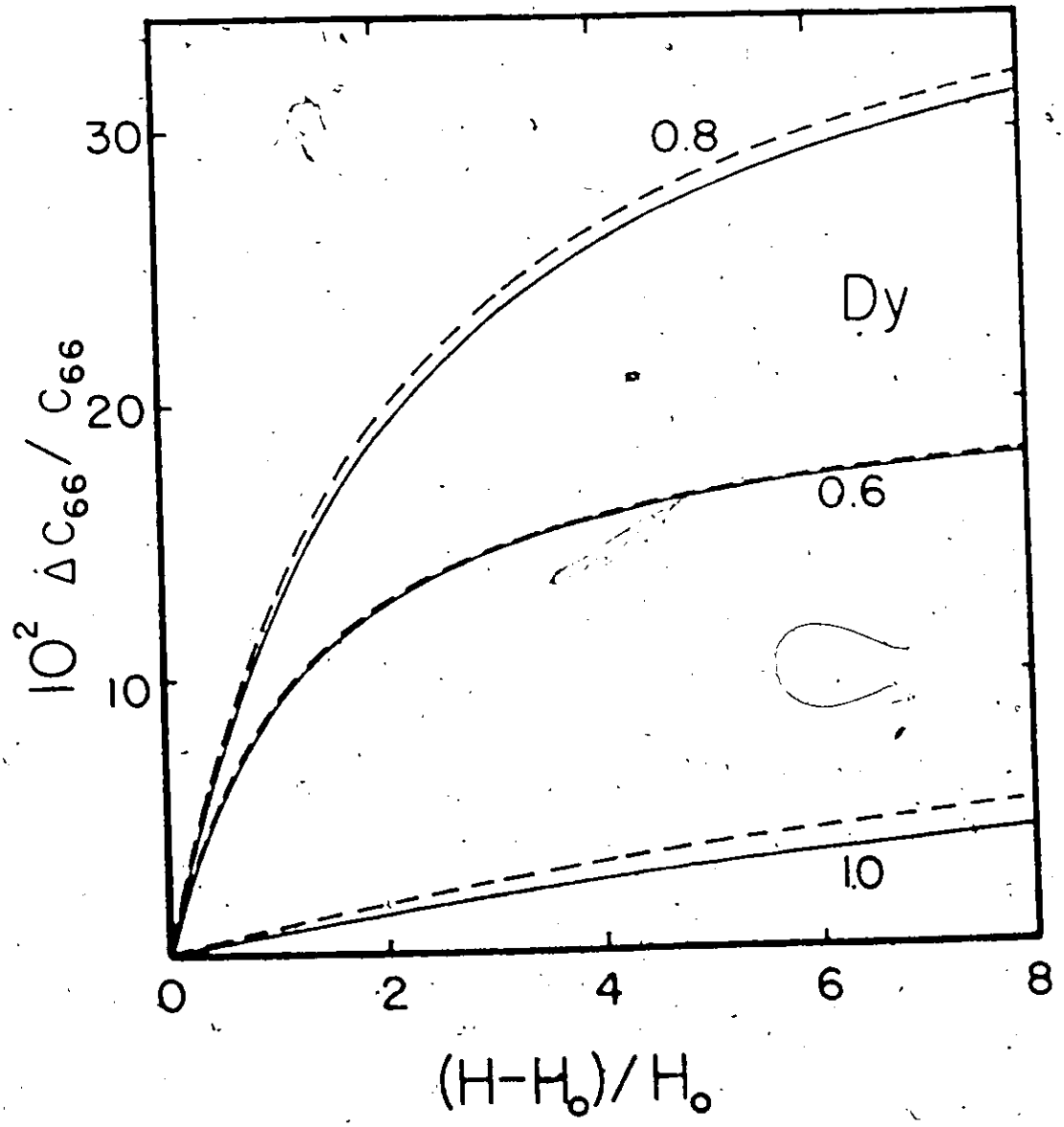

0.75
Tb
20
10
0.5
1.0
0
2
4
6
8
$10^2 \Delta C_{66} / C_{66}$
$(H-H_o)/H_o$

Fig. 6: $\Delta c_{66}/c_{66}$ as a function of $(H - H_0)/H_0$ for Dy calculated from eq. [5.11a] for a magnetic field along the a-axis. $H_0 = 10$ kOe, $b_2^\gamma = 89 \times 10^8$ ergs/cm^3, $b_4^\gamma = 0$ and $c_{66} = 0.263 \times 10^{12}$ ergs/cm^3. Solid curves are for propagation along the b-axis and dashed curves are for propagation along the a-axis. Curves are shown for values of m ranging from 1.0 to 0.6.


30
20
10
0
2
4
6
8
0.8
Dy
0.6
1.0
$10^2\ \Delta C_{66} / C_{66}$
$(H-H_0)/H_0$

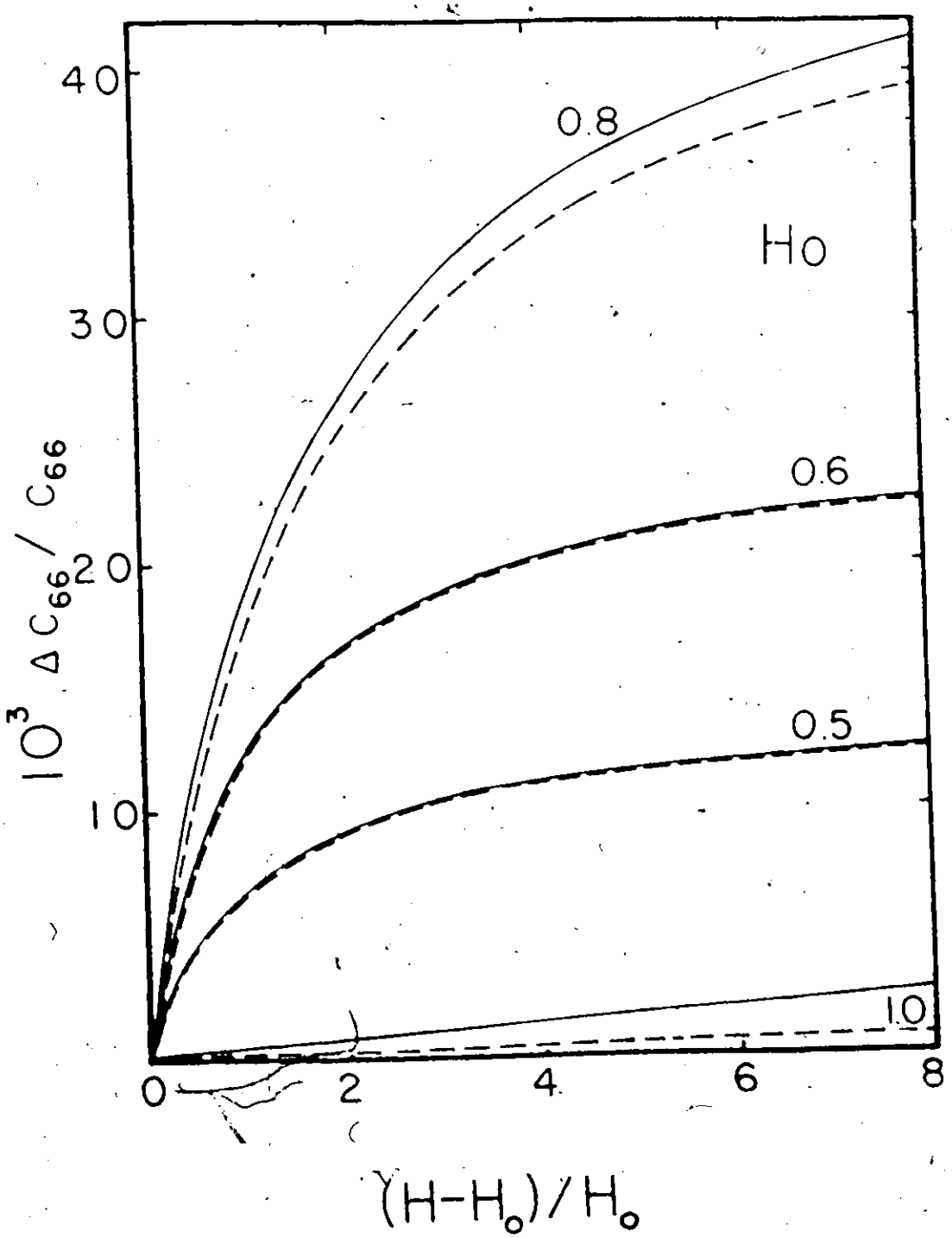
The fact that the largest changes do not occur at the lowest temperatures is due to the denominator of eq. [5.11a] decreasing rapidly as m decreases from 1.0 until, when m is about 0.8, the function H_A becomes comparable with H_0. The curvature of the curves other than m = 1.0 is also due to the function H_A having fallen nearly to zero so that only the magnetic field term is appreciable in the denominator of [5.11a]. In the case of Tb the curve for m = 1.0 is further depressed because the competition between b_2^γ and b_4^γ produces a maximum in H_{ME} near m = 0.92.

Holmium and Erbium

Since there is no magnetoelastic data from which to estimate b_2^ϵ for holmium and erbium it is not possible to predict the size of $\Delta c_{44}/c_{44}$ for these metals. Measurements of appropriate sound velocities should yield reliable estimates of this coupling constant and perhaps of the anisotropy constants as well. It may be seen in Table 1 that there is a considerable difference between the values of b_2 and b_4 for Ho obtained from the work of Feron et al. (1970) and from Bozorth et al. (1968).

In Fig. 7 we have plotted estimates of $\Delta c_{66}/c_{66}$ for Ho based on eq. [5.11] using $b_6^6 = 27 \times 10^6$ ergs/cm^3, the value deduced by Feron et al. (1970). The solid curves are for waves propagating along the a-direction and polarized in

Fig. 7: $\Delta c_{66}/c_{66}$ as a function of $(H - H_0)/H_0$ for Ho calculated from eq. [5.11a] for a magnetic field along the b-axis. H_0 = 10 kOe, $b_2^Y = 31 \times 10^8$ ergs/cm^3, $b_4^Y = 0$ and $c_{66} = 0.308 \times 10^{12}$ ergs/cm^3. Solid curves are for propagation along the a-axis and dashed curves are for propagation along the b-axis. Curves are shown for values of m ranging from 1.0 to 0.5.


40
30
20
10
0
2
4
6
8
0.8
Ho
0.6
0.5
1.0
$10^3 \Delta C_{66} / C_{66}$
$\gamma(H-H_o)/H_o$

the b-direction, while the dashed curves have these two directions interchanged. The magnetic field was taken to be along the easy direction (b-axis) and H_0 was chosen to be 10 kOe. As for Tb and Dy, the greatest change occurs in the region m = 0.75 as a result of H_A falling rapidly to zero with decreasing m in eq. [5.11b]. Rather similar results are obtained for Er using the parameter values given in Tables 1 and 3, but the scale of the curves is increased by a factor of about 3.5. This is mainly due to the fact that b_2^γ is about twice as large for Er compared with Ho.

If the very much smaller value of $b_6^6 = 0.21 \times 10^6$ ergs/cm^3 for Ho due to Bozorth et al. (1968) is used in the calculations, then the function H_A in [5.11b] is almost negligible and the denominator depends almost entirely on the magnetic field terms H and H_0. The result is that the greatest fractional change in c_{66} occurs for m = 1.0, with a value about 3.5 times the maximum in Fig. 7. Thus the magnitude of the change is rather sensitive to the value of b_6^6. When b_6^6 is small, it is also sensitive to the choice of H_0.

5.5 Longitudinal Waves in the Paramagnetic Region

As discussed in Chapters 2 and 3, the requirement of rotational invariance and the use of the finite strain

tensor led to additional magnetoelastic terms linear in the antisymmetric strains $\omega_{\mu\nu}$. For the case of longitudinal sound waves, there occur other terms quadratic in the infinitesimal strains $\epsilon_{\mu\mu}$ which arise entirely from the definition of the finite strains. From eq. [2.20] we have for a pure longitudinal wave,

$$[5.14] \quad E_{\mu\mu} = \epsilon_{\mu\mu} + (\tfrac{1}{2})\ \epsilon_{\mu\mu}^2 \ .$$

When these are retained in the single-ion magnetoelastic Hamiltonian [5.2], with the term in M_{44}^{γ} neglected, and thermal averages of spin operators are taken, we obtain the following expressions for the changes in elastic constants:

$$[5.15a] \quad \Delta c_{33} = - [b_1^{\alpha} + (\tfrac{2}{3}) b_2^{\alpha}] P_2^0(\cos\theta) \langle O_{20} \rangle / [SS(\tfrac{1}{2})]$$

$$[5.15b] \quad \Delta c_{11} = - [b_1^{\alpha} - (\tfrac{1}{3}) b_2^{\alpha}] P_2^0(\cos\theta) \langle O_{20} \rangle / [SS(\tfrac{1}{2})]$$

$$\mp (\tfrac{1}{4}) b_2^{\gamma} \sin^2\theta \cos 2\phi \langle O_{20} \rangle / [SS(\tfrac{1}{2})] \ .$$

where b_1^{α} and b_2^{α} are defined by

$$[5.16a] \quad b_1^{\alpha} = N_a M_{20}^{\alpha,1} SS(\tfrac{1}{2})$$

$$[5.16b] \quad b_2^{\alpha} = N_a M_{20}^{\alpha,2} SS(\tfrac{1}{2}) \ .$$

In eq. [5.15b] the upper sign is for propagation along the a-axis and the lower sign for propagation along the b-axis. The thermal averages $\langle O_{20} \rangle$ are to be taken with respect to the equilibrium spin direction, which is specified by (θ,ϕ). In the ferromagnetic region at low temperatures $\langle O_{20} \rangle / [SS(\frac{1}{2})]$ reduces to m^3 in the usual way (Goodings and Southern 1971). However, in the paramagnetic region when $m(T,H) << 1$, one can use the approximate relation (Callen and Callen 1965; Moran and Lüthi 1970)

$$[5.17] \quad \langle O_{20} \rangle \approx (\tfrac{3}{5}) SS(\tfrac{1}{2}) [m(T,H)]^2$$

Introducing the susceptibility $\chi(T)$ through

$$[5.18] \quad M(T,H) = \chi(T)H$$

we have

$$[5.19] \quad \langle O_{20} \rangle \approx (\tfrac{3}{5}) SS(\tfrac{1}{2}) [\chi(T)/M(T=0)]^2 H^2$$

This is to be substituted in eqs. [5.15a] and [5.15b] to obtain the changes Δc_{33} and Δc_{11} in the paramagnetic region.

The effect of two-ion magnetoelastic terms may be included by considering the Hamiltonian in eq. [5.12] and retaining terms which arise from the quadratic terms in

[5.14]. The changes in elastic constants become

$$[5.20a]\quad \Delta c_{33} = \{-(\tfrac{3}{5})[b_1^\alpha + (\tfrac{2}{3})b_2^\alpha]P_2^0(\cos\theta) - [d_{11}^\alpha + (\tfrac{2}{3})d_{21}^\alpha] - [d_{12}^\alpha + (\tfrac{2}{3})d_{22}^\alpha]P_2^0(\cos\theta)\}[\chi(T)/M(T=0)]^2H^2$$

$$[5.20b]\quad \Delta c_{11} = \{-(\tfrac{3}{5})[b_1^\alpha - (\tfrac{1}{3})b_2^\alpha]P_2^0(\cos\theta) - [d_{11}^\alpha - (\tfrac{1}{3})d_{21}^\alpha] - [d_{12}^\alpha - (\tfrac{1}{3})d_{22}^\alpha]P_2^0(\cos\theta) \mp (\tfrac{1}{4})[(\tfrac{3}{5})b_2^\gamma + d^\gamma]\sin^2\theta\cos 2\phi\}[\chi(T)/M(T=0)]^2H^2$$

where the following definitions have been used

$$[5.21a]\quad d_{11}^\alpha = N_a S^2 \tfrac{1}{2} \sum_j D_{11ij}^\alpha$$

$$[5.21b]\quad d_{21}^\alpha = N_a S^2 \tfrac{1}{2} \sum_j D_{21ij}^\alpha$$

$$[5.21c]\quad d_{12}^\alpha = N_a S^2 \tfrac{1}{2} \sum_j D_{12ij}^\alpha$$

$$[5.21d]\quad d_{22}^\alpha = N_a S^2 \tfrac{1}{2} \sum_j D_{22ij}^\alpha$$

and d^γ is defined in [5.13c]. The summations in [5.21] are over all neighbours of atom i.

The changes in elastic constants in a magnetic field for longitudinal sound waves have been measured for Dy and Ho

in the paramagnetic region by Moran and Lüthi (1970). In both cases they found a quadratic dependence on field outside the critical region which they attributed to terms of second order in the strains $\epsilon_{\mu\mu}$ originating either in the exchange coupling or in single-ion terms and depending on a second derivative of the corresponding coupling constant with respect to strain. The results obtained here from the requirement of finite strains likewise vary as H^2 but depend essentially on a first derivative with respect to strain.

An alternative explanation for the longitudinal elastic constants to that of Moran and Lüthi was proposed by Long et al. (1969). They consider the magnetoelastic coupling as a perturbation carried to second order, within the framework of the usual small strain formulation. This gives results that depend on the square of the magnetoelastic constants instead of linearly as in eq. [5.20].

From the data of Moran and Lüthi for Dy and Ho we can make a fairly accurate estimate of the combination in curly brackets of eq. [5.20a] for $\theta = \pi/2$, assuming that the present explanation dominates other mechanisms. Using the experimental $\chi(T)$ fitted by a Curie-Weiss law, and making use of the relation

$$[5.22] \quad \frac{\Delta c}{c} = \frac{2\Delta v}{v_0}$$

where v_0 is the sound velocity in zero field in the paramagnetic region, we find the values $.20 \times 10^{12}$ ergs/cm^3 for Dy and 2.2×10^{12} ergs/cm^3 for Ho. To obtain these results we used a value of $c_{33} = .787 \times 10^{12}$ ergs/cm^3 for Dy measured by Fisher and Dever (1967) at 300K and a value of $c_{33} = .755 \times 10^{12}$ ergs/cm^3 for Ho from the data of Rosen (1967) at 300K. The value of the two-ion magnetoelastic coupling constant quoted by Pollina and Lüthi (1969) for Dy ($DS^2 = 0.2 \times 10^{12}$ ergs/cm^3) is of the right order of magnitude to account for the value deduced above, but their value for Ho ($DS^2 = .14 \times 10^{12}$ ergs/cm^3) is more than an order of magnitude too small.

From eq. [5.20] it can be seen that it may be possible to deduce separate values for $(\frac{3}{5})b_1^\alpha + d_{12}^\alpha$, $(\frac{3}{5})b_2^\alpha + d_{22}^\alpha$, d_{11}^α, d_{21}^α and $(\frac{3}{5})b_2^\gamma + d^\gamma$ by carrying out measurements for several different directions of the applied field.

CHAPTER 6

HEAVY RARE EARTH-IRON COMPOUNDS RFe_2

The cubic rare earth-iron Laves phase compounds present an interesting possibility for observing large magnetoelastic effects on the measured elastic constants. These compounds have the $MgCu_2$-type structure with the rare earth ions arranged on a diamond sublattice and the iron ions on a corner-sharing network of regular tetrahedra. Recent measurements (Koon et al. 1971; Clark and Belson 1972a, 1972b; Clark et al. 1972) indicate that the magnetic anisotropy and magnetostriction in these compounds is the largest ever observed at room temperature. The magnetic properties of these compounds are well described by a magnetic model in which the rare earth and iron sublattices have their spins directed oppositely and in which the dominant interaction is the exchange term between the two sublattices (Buschow and Stapele 1970; Slanicka et al. 1971; Burzo 1971; Atzmony et al. 1972; Taylor and Darby 1972). If one specializes the general spin-wave theory for a ferrimagnet (Keffer 1966, Sec. 48) to the case where exchange is predominantly between sublattices and where in addition the exchange energy per atom is large compared with the anisotropy energy or Zeeman energy, then there are two spin-wave modes, the lower of which has the

same form as in an ordinary ferromagnet and does not depend on the exchange interactions in the limit of long wavelengths. If we also assume that the magnetostriction and magnetic anisotropy of these compounds is due primarily to the rare earth sublattice (Clark and Belson 1972a, 1972b; Atzmony et al. 1973), then the results obtained in Chapter 4 may be used to predict the size of the effects that may be expected in measurements of the elastic constants as a function of an applied magnetic field. This follows from the fact that the Hamiltonian which describes the coupling of the acoustic magnons to the acoustic phonons has the same form as in eq. [3.38].

6.1 The Hamiltonian for Cubic Symmetry

The Hamiltonian for the rare earth sublattice is taken to consist of a crystalline electric field of cubic symmetry and the Zeeman term:

$$[6.1]\quad \mathcal{H}_m = B_4^0 \sum_j [O_{40}(\underline{S}_j^*) + (\tfrac{10}{7})^{1/2} O_{44}^+(\underline{S}_j^*)] + B_6^0 \sum_j [O_{60}(\underline{S}_j^*) - (14)^{1/2} O_{64}^+(\underline{S}_j^*)] + g\mu_B \sum_j \underline{S}_j^* \cdot \underline{H}^* .$$

The exchange term, which is assumed to be isotropic, has not been included in [6.1] because it does not contribute

to our results in the long wavelength limit. In addition to the terms in the magnetic Hamiltonian in [6.1] we must also include the Zeeman term for the iron sublattice.

Following Callen and Callen (1965), the single-ion magnetoelastic Hamiltonian for the rare earth sublattice is formed by taking products of the symmetry strains and spin functions belonging to the same irreducible representation of the cubic point group. In terms of the spin-operator functions defined in eq. [3.4] it has the form (Southern 1973)

$$[6.2a]\quad \mathcal{H}^I_{me} = \sum_j \mathcal{H}^I_{me}(j)$$

$$[6.2b]\quad \mathcal{H}^I_{me}(j) = - M_0^\alpha E^\alpha - M_4^\alpha E^\alpha [O_{40}(\underset{\sim}{S}_j^q) + (\tfrac{10}{7})^{1/2} O_{44}^+(\underset{\sim}{S}_j^q)]$$

$$- M_2^\gamma \{E_1^\gamma O_{20}(\underset{\sim}{S}_j^q) + E_2^\gamma (2)^{1/2} O_{22}^+(\underset{\sim}{S}_j^q)\}$$

$$- M_4^\gamma \{E_1^\gamma [O_{40}(\underset{\sim}{S}_j^q) - (\tfrac{14}{5})^{1/2} O_{44}^+(\underset{\sim}{S}_j^q)]$$

$$+ E_2^\gamma [- (\tfrac{24}{5})^{1/2} O_{42}^+(\underset{\sim}{S}_j^q)]\}$$

$$- M_2^\epsilon \{E_1^\epsilon [iO_{21}^+(\underset{\sim}{S}_j^q)] + E_2^\epsilon [-iO_{21}^-(\underset{\sim}{S}_j^q)]$$

$$+ E_3^\epsilon [O_{22}^-(\underset{\sim}{S}_j^q)]\}$$

continued...

$$- M_4^{\epsilon}\{E_1^{\epsilon}[i\mathcal{O}_{41}^{+}(S_j^{*}) - (7)^{1/2}\, i\mathcal{O}_{43}^{+}(S_j^{*})]$$

$$+ E_2^{\epsilon}[-i\mathcal{O}_{41}^{-}(S_j^{*}) - (7)^{1/2}\, i\mathcal{O}_{43}^{-}(S_j^{*})]$$

$$+ E_3^{\epsilon}[-(8)^{1/2}\, \mathcal{O}_{42}^{-}(S_j^{*})]\} \quad .$$

Only terms up to $\ell = 4$ have been included. The symmetry strains are defined as follows

[6.3a] $$E^{\alpha} = E_{xx} + E_{yy} + E_{zz}$$

[6.3b] $$E_1^{\gamma} = \frac{(3)^{1/2}}{6}(2E_{zz} - E_{xx} - E_{yy})$$

[6.3c] $$E_2^{\gamma} = \frac{1}{2}(E_{xx} - E_{yy})$$

[6.3d] $$E_1^{\epsilon} = E_{yz}$$

[6.3e] $$E_2^{\epsilon} = E_{xz}$$

[6.3f] $$E_3^{\epsilon} = E_{xy} \quad .$$

The elastic energy associated with the homogeneous strain components may be written in the following form for cubic symmetry

$$[6.4]\quad \mathcal{H}_e = \tfrac{1}{2} c^{\alpha}(E^{\alpha})^2 + \tfrac{1}{2} c^{\gamma}[(E_1^{\gamma})^2 + (E_2^{\gamma})^2] + \tfrac{1}{2} c^{\varepsilon}[(E_1^{\varepsilon})^2 + (E_2^{\varepsilon})^2 + (E_3^{\varepsilon})^2]$$

where the symmetry elastic constants c^{Γ} are related to the usual Cartesian elastic constants as follows

$$[6.5a]\quad c^{\alpha} = \tfrac{1}{3}(c_{11} + 2c_{12})$$

$$[6.5b]\quad c^{\gamma} = 2(c_{11} - c_{12})$$

$$[6.5c]\quad c^{\varepsilon} = 4c_{44} \ .$$

Having ensured that the total angular momentum is conserved we can now carry out the transformation to the usual spin operators S_j and the functions $\varepsilon_{\mu\nu}$ and $\omega_{\mu\nu}$ of small strain theory. We obtain the following new terms linear in $\omega_{\mu\nu}$:

$$[6.6]\quad (20)^{1/2} B_4^0 \sum_j \{\omega_{yz}[(i0_{41}^{+}) + (\tfrac{1}{7})^{1/2}(i0_{43}^{+})] + \omega_{xz}[(-i0_{41}^{-}) - (\tfrac{1}{7})^{1/2}(-i0_{43}^{-})] + \omega_{xy}[-(\tfrac{8}{7})^{1/2} 0_{44}^{-}]\}$$

$$+ (42)^{1/2} B_6^0 \sum_j \{\omega_{yz}[(i0_{61}^{+}) - (\tfrac{5}{2})^{1/2}(i0_{63}^{+}) - (\tfrac{11}{6})^{1/2}(i0_{65}^{+})] + \omega_{xz}[(-i0_{61}^{-}) + (\tfrac{5}{2})^{1/2}(-i0_{63}^{-})]$$

continued...

$$- \left(\frac{11}{6}\right)^{1/2} (-i\bar{\sigma}_{65}^{-})] + \omega_{xy}[\left(\frac{16}{3}\right)^{1/2} \bar{\sigma}_{64}^{-}]\} \quad .$$

In addition to these new linear terms there are also terms quadratic in the displacement gradients. However, since their contributions to the elastic constants are independent of magnetic field, they do not contribute to the results presented in the next section.

6.2 Expressions for Fractional Changes in the Measured Elastic Constants for Cubic Symmetry

Expressions for the changes in the elastic constants due to magnetoelastic interactions are obtained using the methods outlined in Chapters 3 and 4. However, when the Zeeman term for the iron sublattice is included, the expression for the change in the measured elastic constant at two different values of the applied field has a slightly different form than in eq. [4.9]. We have

$$[6.7] \quad c^*(H) - c^*(H_0) = \frac{M_T(H - H_0)[M_R H_{ME} \pm M_R H_A]^2}{4(M_R H_A + M_T H)(M_R H_A + M_T H_0)}$$

where M_T is the total magnetization and M_R is the rare earth sublattice magnetization.

In Table 4 the expressions for $M_R H_{ME}$ and $M_R H_A$ corresponding to the various directions of the magnetization

TABLE 4

EXPRESSIONS FOR MH_A AND MH_{ME}

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}	$K_1+(\frac{1}{2})K_2$	$-2\beta_2-(\frac{5}{2})\beta_5$

are given and, in each case, the elastic constant being measured is indicated. The effective anisotropy field $M_R H_A$ is expressed in terms of the usual macroscopic cubic anisotropy constants K_1 and K_2. These are related to the microscopic constants defined in eq. [6.1] as follows

$$[6.8a] \quad K_1 = -\frac{1}{2}\left[10 b_4 \hat{I}_{9/2} + 21 b_6 \hat{I}_{13/2}\right]$$

$$[6.8b] \quad K_2 = \frac{231}{2} b_6 \hat{I}_{13/2}$$

where

$$[6.8c] \quad b_4 = B_4^0 N_a \, SS(\tfrac{3}{2})$$

$$[6.8d] \quad b_6 = B_6^0 N_a \, SS(\tfrac{5}{2})$$

The temperature dependence has been expressed in the usual way (Callen and Callen 1966) in terms of the (normalized) hyperbolic Bessel function $I_{l+1/2}(\mathcal{L}^{-1}(m_R))$ where $\mathcal{L}^{-1}(m_R)$ is the inverse Langevin function and m_R is the reduced magnetization for the rare earth sublattice. The expressions for $M_R H_{ME}$ involve the magnetoelastic constants of Kittel and Van Vleck (1960) which are related to those defined in eq. [6.2] as follows

$$[6.9a] \quad \beta_0 = -M_0^a N_a$$

[6.9b] $\beta_1 = -\left(\frac{3}{4}\right)^{1/2} M_2^{\gamma} N_a \, SS\left(\frac{1}{2}\right) \hat{I}_{5/2}$

[6.9c] $\beta_2 = -\left(\frac{3}{8}\right)^{1/2} M_2^{\epsilon} N_a \, SS\left(\frac{1}{2}\right) \hat{I}_{5/2}$

[6.9d] $\beta_3 = 5M_4^{\alpha} N_a \, SS\left(\frac{3}{2}\right) \hat{I}_{9/2} - \frac{7(3)^{1/2}}{3} M_4^{\gamma} N_a \, SS\left(\frac{3}{2}\right) \hat{I}_{9/2}$

[6.9e] $\beta_4 = -\frac{7(3)^{1/2}}{2} M_4^{\gamma} N_a \, SS\left(\frac{3}{2}\right) \hat{I}_{9/2}$

[6.9f] $\beta_5 = (5)^{1/2} M_4^{\epsilon} N_a \, SS\left(\frac{3}{2}\right) \hat{I}_{9/2}$

where N_a is the number of rare earth ions per unit volume.

In deriving the expressions in Table 4 for $M_R H_A$, we have not included the contributions of static magnetostriction to K_1 and K_2. At zero temperature these contributions are the order of 1% of K_1 and K_2 and we shall assume that they do not affect the results appreciably.

In each of the cases which we shall consider, the magnetization direction is assumed to be along one of the cubic symmetry directions and the magnetic field is applied in this same direction. In order to minimize domain effects, it is assumed that there is some minimum field strength H_0 which essentially achieves saturation. Only the results for transverse elastic waves which are either propagating or polarized along the magnetization direction are given.

6.3 Estimates for the Compounds RFe_2 (R = Tb, Dy, Ho, Er and Tm)

At the present time single crystal data for the magnetostriction and magnetic anisotropy constants is available only for $ErFe_2$. However, polycrystalline data may be used to estimate the size of the effects to be expected in measurements of the elastic constants as a function of applied field. In making our estimates we shall consider only the lowest order magnetoelastic constants β_1 and β_2 and when single crystal data is not available we shall use the polycrystalline results and make the rather crude approximation of isotropic magnetostriction. In this case

$$[6.10] \quad \beta_1 = \beta_2 = -2\mu\left(\tfrac{3}{2}\right)\lambda_s$$

where μ is the shear modulus of elasticity and λ_s is the saturation magnetostriction. If single crystal data is available then β_1 and β_2 may be determined from the following relations:

$$[6.11a] \quad \beta_1 = -3\mu\lambda_{100}$$

$$[6.11b] \quad \beta_2 = -3\mu\lambda_{111}$$

where λ_{100} and λ_{111} are the usual magnetostriction constants for cubic symmetry.

In Table 5 the values of the various constants required to make our predictions are given at 0°K. The magnetic anisotropy constants b_4 and b_6 have been obtained using [6.8] by comparing the Mössbauer data of Atzmony et al. (1973) with the torque measurements of Clark et al. (1972). Our values are approximately five times larger than those of Atzmony et al. (1973). Values of the shear modulus μ for $TbFe_2$ and $ErFe_2$ were taken from the room temperature data of Clark and Belson (private communication) and the other values were obtained by interpolation based on the room temperature values for the rare earth metals. The magnetoelastic coupling constants β_1 and β_2 for all the RFe_2 compounds except $ErFe_2$ were determined from the data of Clark and Belson (1972a, 1972b) using eq. [6.10]. In the case of $ErFe_2$ the measurements of Clark and Belson (private communication) were used in eq. [6.11] assuming that $\lambda_{100} \simeq (0.1)\lambda_{111}$. The saturation moments were taken in all cases from the data of Burzo (1971).

In Figs. 8 - 12 we have plotted $[c^*(H) - c^*(H_0)]/c^*$ as a function of the applied field and the rare earth sublattice reduced magnetization m_R. The temperature dependence has been introduced using the (normalized) hyperbolic Bessel function $\hat{I}_{L+1/2}(\mathcal{L}^{-1}(m_R))$ and it is assumed

TABLE 5

VALUES OF MAGNETIC CONSTANTS AT 0°K FOR RFe_2 COMPOUNDS

	N_a (10^{22} atoms/cc)	b_4 (10^6 ergs/cc)	b_6 (10^6 ergs/cc)	μ^a (10^{11} ergs/cc)	β_1 (10^9 ergs/cc)	β_2 (10^9 ergs/cc)	M_R (μ_B)	M_{Fe} (μ_B)	M_T (μ_B)
$TbFe_2$	2.02	156.	6.26	3.53	-3.60	-3.60	8.4	1.7	5.0
$DyFe_2$	2.04	-196.	-24.2	4.0	-3.92	-3.92	9.4	1.7	6.0
$HoFe_2$	2.06	-127.	46.8	4.1	-1.53	-1.53	9.4	1.7	6.0
$ErFe_2$	2.08	121.	-35.7	4.62	0.24	2.44	8.4	1.7	5.0
$TmFe_2$	2.10	152.	18.1	4.6	3.96	3.96	6.1	1.7	2.7

a room temperature values

that the iron sublattice magnetization is independent of temperature, as appears to be a reasonable approximation below room temperature (Clark and Belson 1972a; Bargouth and Will 1971; Burzo, private communication). For each compound it is assumed that the applied field is along the easy direction and that a minimum field strength of 10 kOe is sufficient to saturate the magnetization. In each case the solid (dashed) curve corresponds to a transverse elastic wave propagating (polarized) along the easy direction.

As can be easily seen from the figures, the fractional change in elastic constant $\Delta c/c$ is of the order of 10^{-2} at room temperature for each compound. In addition, the difference in $\Delta c/c$ for the two types of transverse wave is the order of 10^{-3}. Thus it should be possible to obtain accurate values of the various magnetoelastic constants and anisotropy constants by carefully analyzing measurements of $\Delta c/c$ as a function of an applied field and temperature using [6.7] and available magnetization data.

Fig. 8: $\Delta c/c$ as a function of $(H - H_0)/H_0$ for $TbFe_2$ with the magnetic field along [111] and H_0 = 10 kOe. Solid (dashed) curves are for propagation (polarization) along [111] and $c = \frac{1}{3}(c_{11} - c_{12} + c_{44})$.

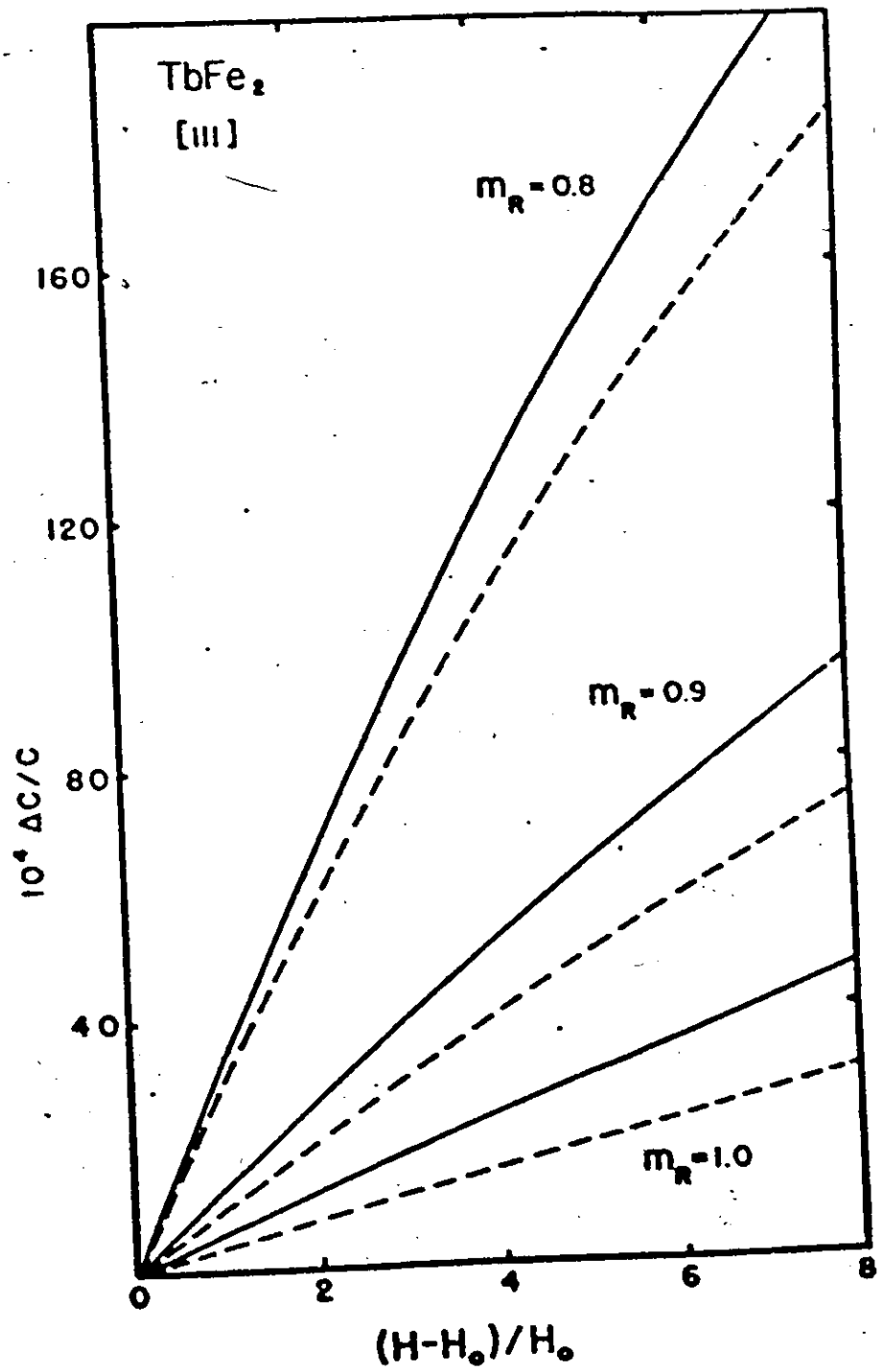

TbFe₂
[111]
m_R = 0.8
m_R = 0.9
m_R = 1.0
160
120
80
40
0
2
4
6
8
10⁴ ΔC/C
(H-H₀)/H₀

Fig. 9: $\Delta c_{44}/c_{44}$ as a function of $(H - H_0)/H_0$ for $DyFe_2$ with the magnetic field along [001] and H_0 = 10 kOe. Solid (dashed) curves are for propagation (polarization) along [001].

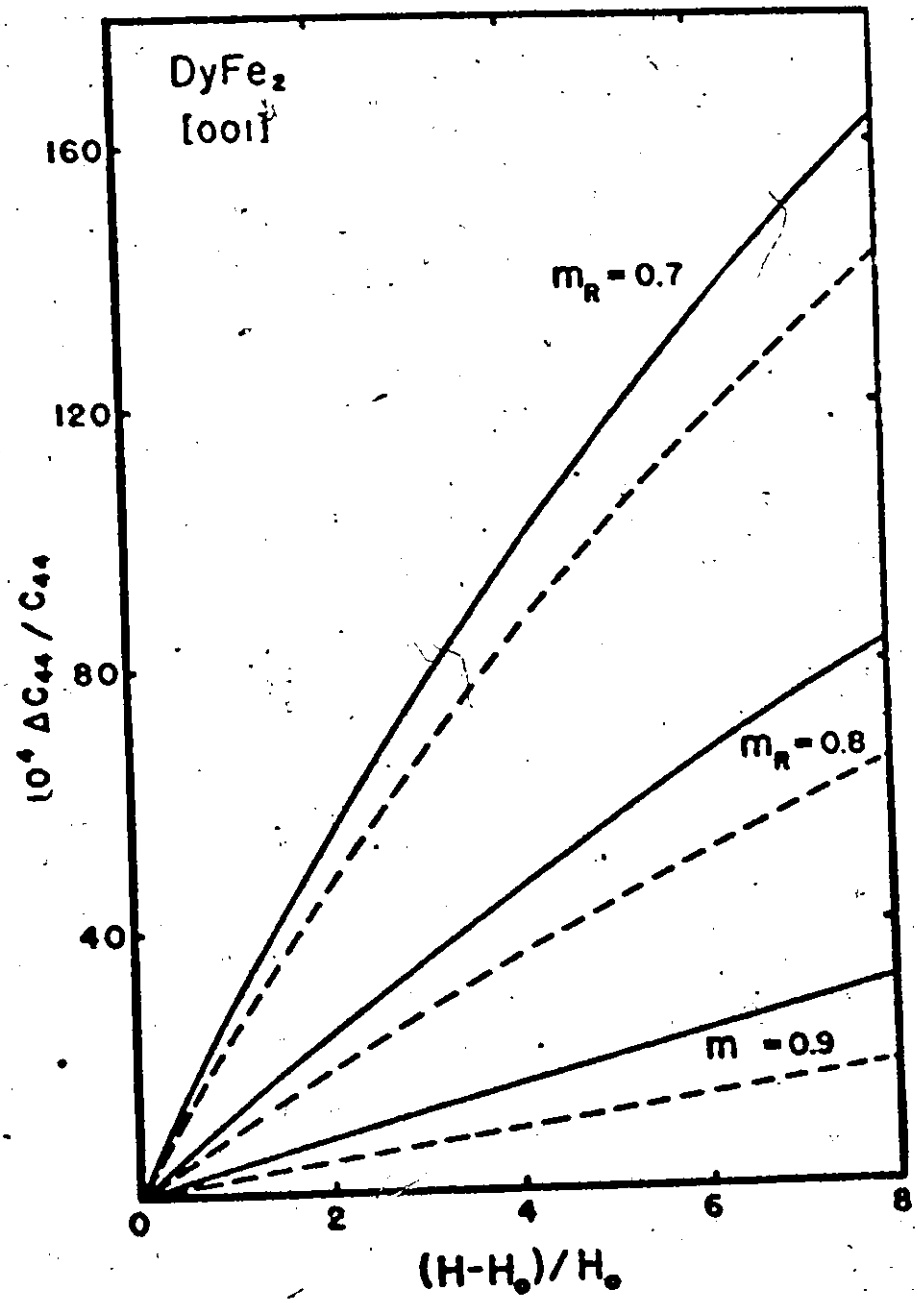

DyFe2
[001]
160
120
80
40
0
2
4
6
8
mR = 0.7
mR = 0.8
m = 0.9
10⁴ ΔC44 / C44
(H-H₀)/H₀

Fig. 10: $\Delta c_{44}/c_{44}$ as a function of $(H - H_0)/H_0$ for $HoFe_2$ with the magnetic field along [001] and H_0 = 10 kOe. Solid (dashed) curves are for propagation (polarization) along [001].

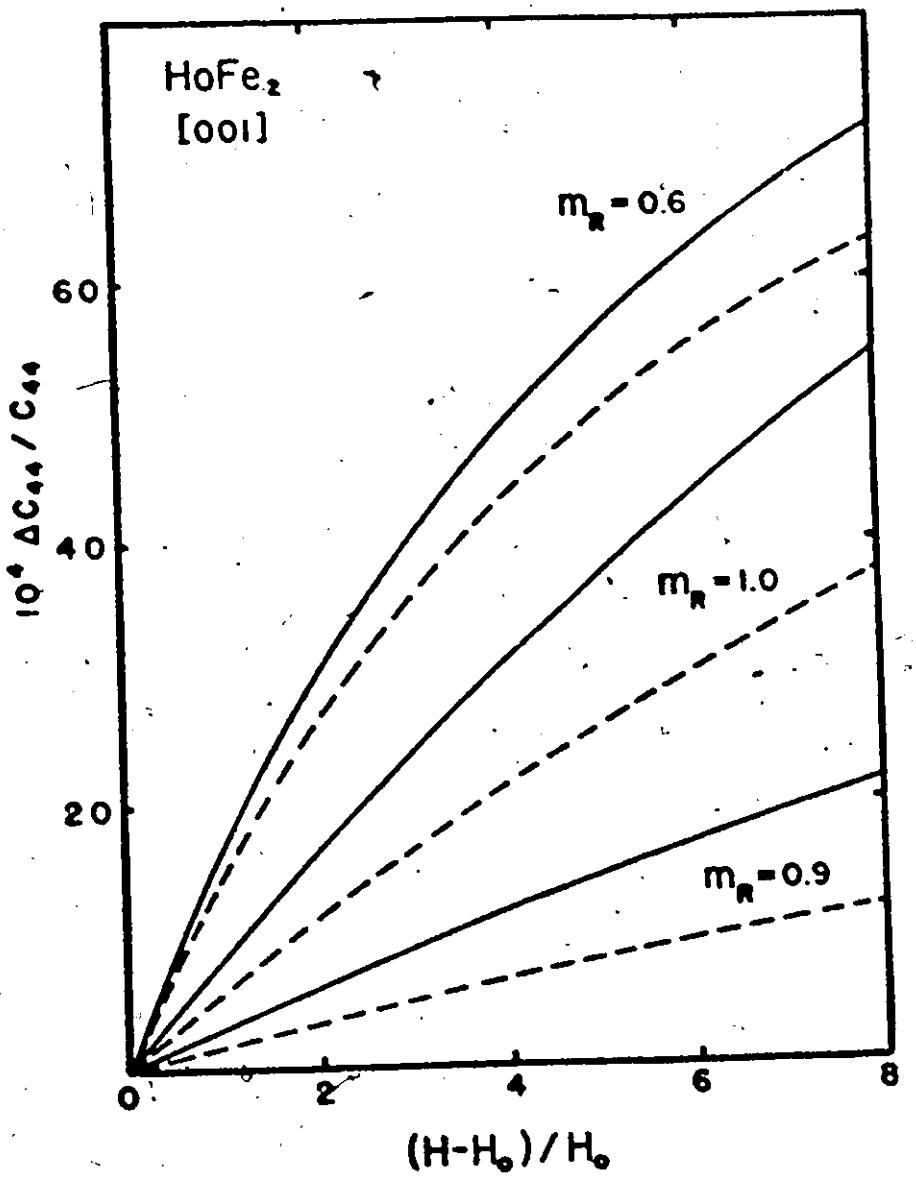

HoFe2
[001]
m_R = 0.6
m_R = 1.0
m_R = 0.9
60
40
20
0
2
4
6
8
10⁴ ΔC44 / C44
(H-H0) / H0

Fig. 11: $\Delta c/c$ as a function of $(H - H_0)/H_0$ for $ErFe_2$ with the magnetic field along [111] and H_0 = 10 kOe. Solid (dashed) curves are for propagation (polarization) along [111] and $c = \frac{1}{3}(c_{11} - c_{12} + c_{44})$.

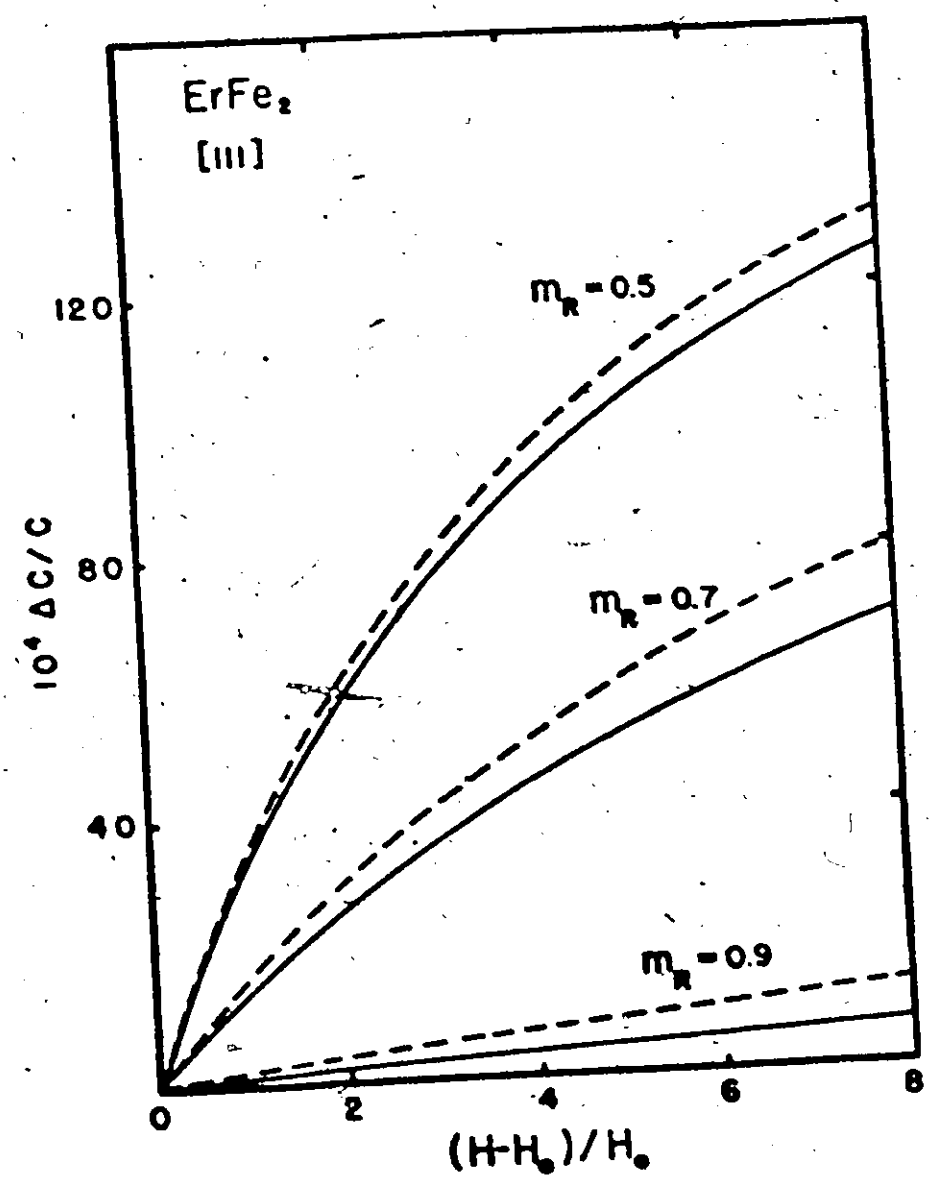
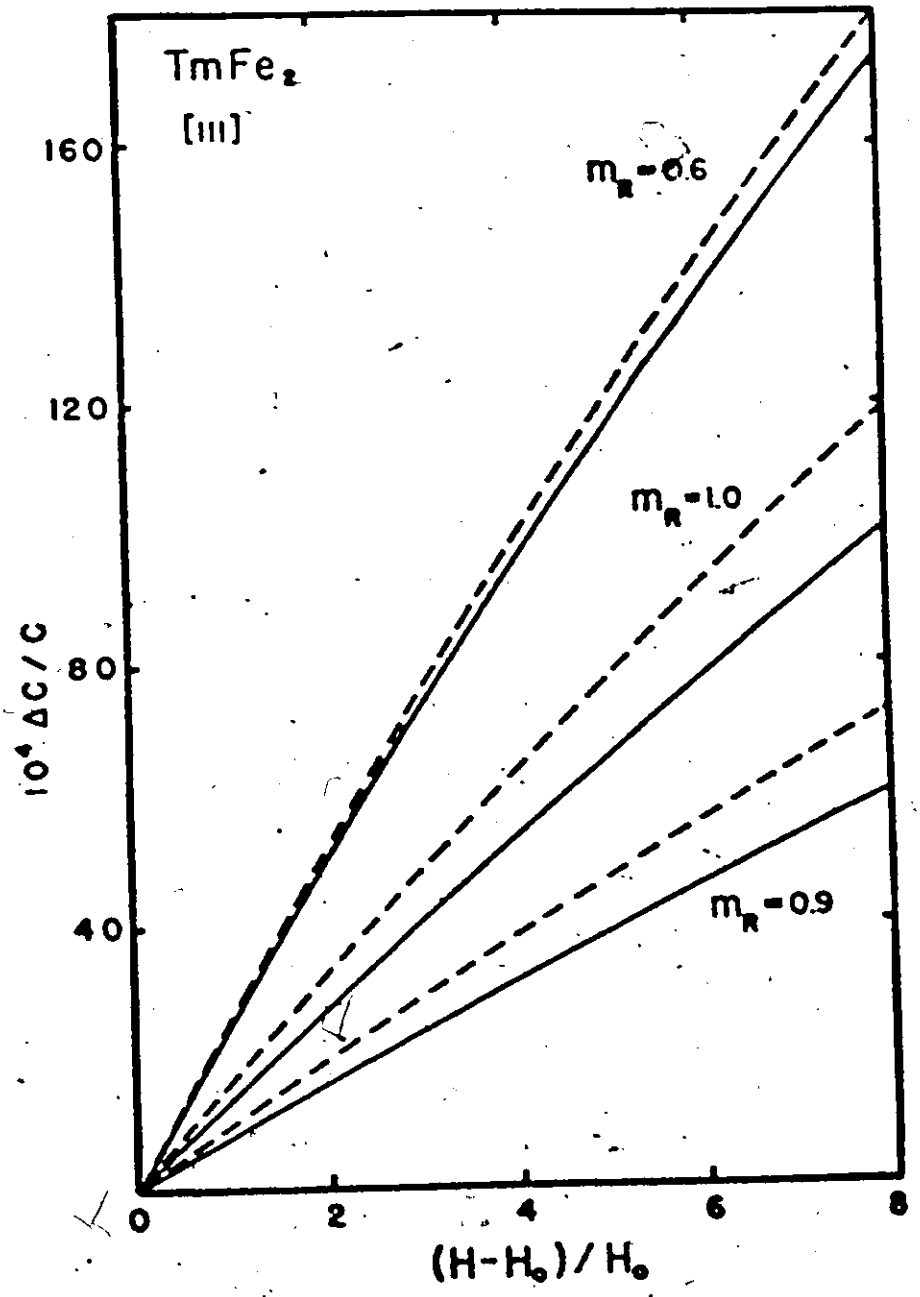

ErFe₂
[111]
m_R = 0.5
m_R = 0.7
m_R = 0.9
120
80
40
0
2
4
6
8
$10^4 \Delta C/C$
$(H-H_0)/H_0$

Fig. 12: $\Delta c/c$ as a function of $(H - H_0)/H_0$ for $TmFe_2$ with the magnetic field along [111] and H_0 = 10 kOe. Solid (dashed) curves are for propagation (polarization) along [111] and $c = \frac{1}{3}(c_{11} - c_{12} + c_{44})$.


TmFe2
[111]
160
120
80
40
10⁴ ΔC / C
0
2
4
6
8
(H-H₀) / H₀
m_R=0.6
m_R=1.0
m_R=0.9

CHAPTER 7

SUMMARY

A rotationally invariant theory of magnetoelastic coupling in ferromagnets has been presented in this thesis. Both the static and dynamic effects of this coupling have been discussed. The static coupling, or magnetostriction, gives rise to static distortions of the crystal which depend upon the equilibrium moment direction. In addition, this static coupling also contributes to the magnetic anisotropy in determining the equilibrium moment configuration. The dynamic magnetoelastic coupling leads to coupled oscillations in the magnetic and elastic degrees of freedom about the equilibrium configuration. Previous treatments of this dynamic interaction in ferromagnets appear to be incorrect. In particular, they neglect an additional coupling which originates in the magnetic anisotropy of the crystal and involves the antisymmetric strain functions $\omega_{\mu\nu}$. In the case of transverse elastic waves which are either propagating or polarized along the direction of magnetization, these new linear terms result in new effects similar to those found by Melcher (1970) in MnF_2, from which it should be possible to obtain in a direct manner the values of certain magnetoelastic constants and anisotropy constants.

Expressions for the change in the measured elastic constant for transverse elastic waves at two different values of an applied magnetic field have been derived. The quantities appearing in these expressions have been described in terms of macroscopic anisotropy constants and magnetostriction constants for both hexagonal-close-packed and cubic symmetry. These expressions can be used in the analysis of measurements of the changes in elastic constant as a function of the applied field strength and temperature.

Using available data for the magnetoelastic constants and the anisotropy constants, this process has been turned around to estimate the size of the effects which can be expected to be measured experimentally in the case of the heavy rare earth metals and some of their compounds. It is predicted that fractional changes in the measured elastic constants as large as 10^{-2} should be found for Tb, Dy, Ho and Er in fields of about 50 kOe, while the maximum change for Gd is predicted to be about 10^{-4}. Similarly, the fractional change in elastic constant for the rare earth-iron compounds RFe_2 (R = Tb, Dy, Ho, Er and Tm) is predicted to be as large as 10^{-2} at room temperature for each compound. Thus it should be possible to obtain accurate values for the various magnetoelastic constants and anisotropy constants by carefully analyzing the measurements as a function of an applied field and temperature using the appropriate expressions.

Calculations have also been performed for the field-dependent changes in c_{11} and c_{33} for longitudinal waves in the paramagnetic region. These changes result from the fact that the finite strains $E_{\mu\mu}$ include terms of the form $\epsilon^2_{\mu\mu}$. The resulting changes in c_{11} and c_{33} depend linearly on the magnetoelastic constants and vary as H^2 in the paramagnetic region. Estimates of certain combinations of these constants have been made from the experimental measurements of Moran and Lüthi on Dy and Ho.

APPENDIX A

ENERGY OF THE UNIFORM-MAGNON MODE IN THE PRESENCE OF MAGNETOELASTIC COUPLING

The energy of the $\underline{q} = 0$, or uniform-magnon, mode in a highly anisotropic ferromagnet may be obtained using the macroscopic theory developed by Smit and Belgers (1955). Denoting the second derivatives of the macroscopic energy density as $F_{\alpha\beta}$, they obtain the following expression for the uniform-mode energy

$$[A.1] \qquad E_{\underline{q}=0} = \frac{gu_B}{M(T)\sin\theta} [F_{\theta\theta}F_{\phi\phi} - (F_{\theta\phi})^2]^{1/2}$$

where the derivatives $F_{\alpha\beta}$ are to be evaluated at the equilibrium values of the polar angles θ and ϕ specifying the direction of spontaneous magnetization. The equilibrium values are determined from the conditions $\partial F/\partial\alpha = 0$ ($\alpha = \theta, \phi$). If the magnetization direction coincides with a crystal symmetry direction, then $F_{\theta\phi}$ vanishes and [A.1] may be written simply as

$$[A.2] \qquad E_{\underline{q}=0} = gu_B[H_A^{\theta}H_A^{\phi}]^{1/2}$$

where

$$H_A^{\theta} = \frac{1}{M(T)} F_{\theta\theta}$$

[A.3]

$$H_A^{\phi} = \frac{1}{M(T)\sin^2\theta} F_{\phi\phi} \quad .$$

These latter quantities represent the effective anisotropy fields acting on the spin components when they depart from the equilibrium direction. In a dynamic experiment such as ferromagnetic resonance, the uniform-magnon mode is excited and these effective fields can be measured.

The effective anisotropy field measured in the static torque and magnetization experiments may be identified by comparing the energy density F with the potential energy of a magnetization vector $\underset{\sim}{M}$ in a magnetic field $\underset{\sim}{H}$. In this case

[A.4] $$F = - \underset{\sim}{M}.\underset{\sim}{H} = - MH \cos\alpha$$

where α denotes the angle between $\underset{\sim}{M}$ and $\underset{\sim}{H}$. Evaluating $F_{\alpha\alpha}$ at the equilibrium value $\alpha = 0$, we have

[A.5] $$F_{\alpha\alpha} = MH \cos\alpha\Big|_{\alpha=0} = MH \quad .$$

The effective anisotropy field may be identified as

[A.6] $$H_A^{\alpha} = \frac{1}{M(T)} F_{\alpha\alpha}$$

where $F_{\alpha\alpha}$ is to be evaluated at the equilibrium value of α. Note that this equation agrees with [A.3] provided both the magnetization and the effective anisotropy field are correctly projected onto the plane in which the angle ϕ is measured. The effective anisotropy field measured in the dynamic and static experiments appear to be the same and may be expressed in terms of macroscopic quantities using an expression such as [A.6].

The above results are usually assumed to remain valid in systems where magnetoelastic interactions are present. However, it has become apparent that the effective anisotropy fields measured in the static and dynamic experiments are not the same when magnetoelastic coupling is taken into account. In the past several years, there has been a great deal of theoretical and experimental work performed to determine the effect of magnetoelastic coupling on the uniform-magnon modes in several of the heavy rare earth metals. Turov and Shavrov (1965) suggested that the correct way to find the uniform-mode energy is to regard the strain as frozen at its equilibrium position. Then, when a long-wavelength magnon is excited, the lattice strain does not follow the precession of the spin components. Cooper (1968a), using this frozen-lattice model, found that there is a significant magnetoelastic contribution to the uniform-mode energy.

Cooper also examined the case in which the lattice strain can follow the spin precession and found that there is also a magnetoelastic contribution in this case. This so-called free-lattice model predicts a much smaller value for the uniform-mode energy. In order to compare the results predicted by these two models, the contributions of magnetoelastic coupling to the magnetic anisotropy must be properly extracted in each case.

As an example, consider the following form for the energy density

$$[A.7] \quad F = K(\theta,\phi) + b(\theta,\phi)E^{\Gamma} + \frac{1}{2} c^{\Gamma}(E^{\Gamma})^2$$

where the first term represents the usual magnetic anisotropy energy. The latter terms represent the magnetoelastic and elastic energies respectively for the irreducible representation labelled by Γ. The equilibrium configuration is determined by minimizing F with respect to θ, ϕ and E^{Γ}:

$$[A.8a] \quad \frac{\partial F}{\partial E^{\Gamma}} = b(\theta,\phi) + c^{\Gamma}E^{\Gamma} = 0$$

$$[A.8b] \quad \frac{\partial F}{\partial \alpha} = \frac{\partial K}{\partial \alpha} + \frac{\partial b}{\partial \alpha} E^{\Gamma} = 0 \qquad (\alpha = \theta, \phi) \quad .$$

These equations must be solved simultaneously to obtain the equilibrium values θ_0, ϕ_0 and E^{Γ}.

In a dynamic experiment such as ferromagnetic resonance, the equilibrium moment direction is unchanged and the strains remain frozen at their equilibrium values. When using an expression such as [A.6] to obtain the effective anisotropy field, we do not differentiate the strains E^{Γ} with respect to the angles θ and ϕ. The energy density becomes.

$$[A.9] \quad F = K(\theta,\phi) + b(\theta,\phi)\bar{E}^{\Gamma}(\theta_0,\phi_0) + \frac{1}{2} c^{\Gamma}[\bar{E}^{\Gamma}(\theta_0,\phi_0)]^2$$

and the effective anisotropy field is simply

$$[A.10] \quad H_A^{\alpha} = \frac{1}{M}\left[\frac{\partial^2 K}{\partial \alpha^2} + \frac{\partial^2 b}{\partial \alpha^2}\bar{E}^{\Gamma}\right] \qquad (\alpha = \theta, \phi) \quad .$$

On the other hand, in a static experiment such as a magnetic torque measurement, the equilibrium moment direction changes under the influence of an externally applied magnetic field. The strains E^{Γ} follow the direction of the moments and must be differentiated with respect to the angles θ and ϕ when evaluating the effective anisotropy field. However, this is equivalent to substituting the following expression for the strains directly into the energy density:

$$[A.11a] \quad E^{\Gamma} = - b(\theta,\phi)/c^{\Gamma} \quad .$$

We obtain

$$[A.11b]\quad F = K - b^2/c^\Gamma + b^2/2c^\Gamma = K - b^2/2c^\Gamma$$

and the effective anisotropy field in this case is

$$[A.12]\quad H_A^\alpha = \frac{1}{M}\left[\frac{\partial^2 K}{\partial\alpha^2} - \frac{b}{c^\Gamma}\frac{\partial^2 b}{\partial\alpha^2} - \left(\frac{\partial b}{\partial\alpha}\right)^2/c^\Gamma\right]$$

$$= \frac{1}{M}\left[\frac{\partial^2 K}{\partial\alpha^2} + E^\Gamma\frac{\partial^2 b}{\partial\alpha^2} - c^\Gamma\left(\frac{\partial E^\Gamma}{\partial\alpha}\right)^2\right] \qquad (\alpha = \theta,\ \phi)\ .$$

Comparing [A.10] and [A.12], we see that the effective anisotropy field measured in the static experiments is smaller than that measured in the dynamic experiments. The difference is given by

$$[A.13]\quad \Delta H_A^\alpha = H_A^\alpha(\text{dynamic expt}) - H_A^\alpha(\text{static expt})$$

$$= \frac{c^\Gamma}{M}\left(\frac{\partial E^\Gamma}{\partial\alpha}\right)^2\ .$$

In the case of hexagonal-close-packed symmetry, we have for $\theta_0 = \pi/2$ and $\phi = \phi_0$:

$$[A.14a]\quad \Delta H_A^\theta = \frac{c^\varepsilon}{M}\left(\frac{H}{2}\right)^2$$

$$[A.14b]\quad \Delta H_A^{\phi} = \frac{4c^{\gamma}}{M}\,[A^2 + C^2 + 2AC\cos 6\phi_0]$$

where the magnetoelastic coupling constants A, C and H are those defined by Mason (1954). These results agree with those obtained by Brooks (1972).

A great deal of experimental work has been performed to determine whether the frozen-lattice or free-lattice model is correct in describing the dynamic experiments. Cooper was the first to demonstrate that, if a magnetic field is applied along a magnetically hard direction in the basal plane of the hexagonal-close-packed structure, then the two models yield quite different results. In the case of the frozen-lattice model, the uniform-mode energy can be reduced to a minimum nonzero value when the strength of the applied field is equal to the effective planar anisotropy field. On the other hand, the free-lattice model predicts that the energy gap may be reduced to zero at the same value of the applied field. Using neutron-diffraction techniques, Nielsen et al. (1970b) found that the observed gap could not be reduced to zero in the case of Tb and they concluded that the frozen-lattice model was valid. Other groups (Bagguley and Liesegang 1967; Rossol and Jones 1966; Hart and Stanford 1971) have studied this same effect using ferromagnetic resonance techniques. At microwave frequencies far below the minimum spin-wave gap predicted by the frozen-lattice

model, they observe a strong magnon absorption which is clearly inconsistent with the frozen-lattice model. In contrast, investigations of the temperature dependence of the resonance field at higher microwave frequencies (Wagner and Stanford 1969, 1972; Marsh and Sievers 1969) show agreement with the frozen-lattice model.

The experimental observations appear to be quite contradictory and do not indicate which model is the correct one. In general, the results of neutron-diffraction and high-frequency resonance absorption experiments conclude that the frozen-lattice model is valid, whereas the low-frequency resonance absorption cannot be satisfactorily explained using this model.

Vigren and Liu (1972) have attempted to unify the free-lattice and frozen-lattice models. Using a model in which the crystal strains are locally coupled to the spin, they predict the free-lattice model behaviour at low microwave frequencies and the frozen-lattice model behaviour at high microwave frequencies. An alternative explanation for the magnetic absorption observed in the low-frequency microwave experiments has been presented by Chow and Keffer (1973). These authors propose that the results of both ferromagnetic resonance and neutron scattering experiments should be analyzed using the true magnetoelastic modes of the coupled system. All of the experiments performed so far

have been analyzed only in terms of either the free-lattice or frozen-lattice models. Chow and Keffer have demonstrated that under the conditions in which the resonance experiments are performed, that is, with a magnetic field applied along the hard planar direction and equal to the effective planar anisotropy field, neither of these models is adequate for a description of the normal modes of the system. The frozen-lattice model correctly gives the energy of the uniform-magnon mode, but it does not take account of the dynamic coupling between magnons and phonons at finite wavevectors. If there exists a strong coupling between the unperturbed magnon and phonon modes, then the correct normal modes are really magnetoelastic modes and involve coupled spin deviations and lattice displacements. In the case of transverse acoustic phonons which are both propagating and polarized in the basal plane, the dynamic magnetoelastic coupling causes these modes to have a strong magnon character near $q = 0$. In fact, the energy of these modes varies as q^2 in this region in contrast to the usual linear dependence on q. Since the electromagnetic field interacts only with the spin, the contribution of these modes to the magnetic absorption depends on the degree of spin contribution to these modes. Chow and Keffer point out that this fact makes the analysis of the absorption a complicated problem, but they are able to deduce some important features of the

expected behaviour of the absorption as a function of the applied dc magnetic field. They conclude that the results of the low-frequency experiments mentioned previously are consistent with the existence of true magnetoelastic modes in these metals. We believe that this explanation of the resonance results is correct and that the so-called free-lattice model should be abandoned.

APPENDIX B

EVALUATION OF H_{ME}

In Chapter 4 we obtained a simple expression for the change in the measured elastic constant at two different values of an applied magnetic field for transverse elastic waves which are either propagating or polarized along the direction of the magnetization. If α denotes the angle measured from the direction of magnetization in the plane defined by the propagation and polarization directions of the particular transverse elastic wave being considered, then the quantity H_A appearing in [4.9] may be obtained using [A.6]. This quantity is the effective anisotropy field measured in dynamic experiments such as ferromagnetic resonance.

A similar procedure may be used to obtain the quantity H_{ME} appearing in [4.9]. If the elastic constant c^Γ is being measured, then H_{ME} is given by

$$[B.1] \qquad H_{ME} = \frac{c^\Gamma}{M} \frac{\partial E^\Gamma}{\partial \alpha}$$

where the derivative is to be evaluated at the equilibrium value of α and the strain function E^Γ is obtained by minimizing the magnetoelastic and elastic energies as in eq. [A.8a].

As an example, we have for the case of hexagonal-close-packed symmetry the following expressions for the symmetry strains E^{Γ}:

[B.2a] $$E_1^{\varepsilon} = \frac{H}{4} \sin 2\theta \sin\phi$$

[B.2b] $$E_2^{\varepsilon} = \frac{H}{4} \sin 2\theta \cos\phi$$

[B.2c] $$E_2^{\gamma} = C \sin^2\theta \sin 2\phi + \frac{A}{2} \sin^4\theta \sin 4\phi \quad .$$

In a measurement of $c^{\varepsilon} = 4c_{44}$, the angle α is described by θ and we have

[B.3] $$MH_{ME} = c^{\varepsilon} \frac{H}{2} \cos 2\theta \qquad (\phi = 0 \text{ or } \tfrac{\pi}{2})$$

in agreement with eqs. [5.9c] and [5.10c]. In the case of $c^{\gamma} = 4c_{66}$, the angle α is described by ϕ and we obtain for $\theta = \pi/2$

[B.4] $$MH_{ME} = 2c^{\gamma}[C \cos 2\phi + A \cos 4\phi]$$

in agreement with [5.11c].

APPENDIX C

EXPRESSIONS FOR $|V_{q\lambda}|^2$

In this appendix expressions are given for $|V_{q\lambda}|^2$ in terms of the various magnetoelastic and anisotropy constants defined in Eqs. [5.8] and [5.16]. The direction of magnetization is specified by the angles θ and ϕ. As is implied by the notation $V_{q\lambda}$ the first subscript gives the direction of propagation of the sound wave while the second specifies its polarization.

$$|V_{aa}|^2 = \frac{E_q |q|^2 \hbar}{4\rho\omega_{q\lambda}} \left\{ \frac{[(6b_1^\alpha m^3 - 2b_2^\alpha m^3 - b_c^\gamma(\theta))\sin 2\theta]^2}{16N_a Sm(A_0+B_0)} + \frac{[b_s^\gamma(\theta)\sin\theta]^2}{4N_a Sm(A_0-B_0)} \right\}$$

$$|V_{bb}|^2 = \frac{E_q |q|^2 \hbar}{4\rho\omega_{q\lambda}} \left\{ \frac{[(6b_1^\alpha m^3 - 2b_2^\alpha m^3 + b_c^\gamma(\theta))\sin 2\theta]^2}{16N_a Sm(A_0+B_0)} + \frac{[b_s^\gamma(\theta)\sin\theta]^2}{4N_a Sm(A_0-B_0)} \right\}$$

$$|V_{cc}|^2 = \frac{E_q |q|^2 \hbar}{4\rho\omega_{q\lambda}} \left\{ \frac{[(3b_1^\alpha + 2b_2^\alpha) m^3 \sin 2\theta]^2}{4N_a Sm(A_0+B_0)} \right\}$$

$$|V_{ca}|^2 = \frac{E_q |q|^2 \hbar}{4\rho\omega_{q\lambda}} \left\{ \frac{[b_2^{\epsilon} m^3 \cos 2\theta + f_2(\theta)]^2 \cos^2\phi}{4N_a Sm(A_0+B_0)} + \frac{[b_2^{\epsilon} m^3 + f_3(\theta)]^2 \cos^2\theta \sin^2\phi}{4N_a Sm(A_0-B_0)} \right\}$$

$$|V_{ac}|^2 = \frac{E_q |q|^2 \hbar}{4\rho\omega_{q\lambda}} \left\{ \frac{[b_2^{\epsilon} m^3 \cos 2\theta - f_2(\theta)]^2 \cos^2\phi}{4N_a Sm(A_0+B_0)} + \frac{[b_2^{\epsilon} m^3 - f_3(\theta)]^2 \cos^2\theta \sin^2\phi}{4N_a Sm(A_0-B_0)} \right\}$$

$$|V_{cb}|^2 = \frac{E_q |q|^2 \hbar}{4\rho\omega_{q\lambda}} \left\{ \frac{[b_2^{\epsilon} m^3 \cos 2\theta + f_2(\theta)]^2 \sin^2\phi}{4N_a Sm(A_0+B_0)} + \frac{[b_2^{\epsilon} m^3 + f_3(\theta)]^2 \cos^2\theta \cos^2\phi}{4N_a Sm(A_0-B_0)} \right\}$$

$$|V_{bc}|^2 = \frac{E_q |q|^2 \hbar}{4\rho\omega_{q\lambda}} \left\{ \frac{[b_2^{\epsilon} m^3 \cos 2\theta - f_2(\theta)]^2 \sin^2\phi}{4N_a Sm(A_0+B_0)} + \frac{[b_2^{\epsilon} m^3 - f_3(\theta)]^2 \cos^2\theta \cos^2\phi}{4N_a Sm(A_0-B_0)} \right\}$$

$$|V_{ba}|^2 = \frac{E_q|q|^2\hbar}{4\rho\omega_{q\lambda}}\left\{\frac{[b_s^\gamma(\theta)\sin2\theta]^2}{16N_aSm(A_0+B_0)} + \frac{[b_c^\gamma(\theta)+f_3(\theta)]^2\sin^2\theta}{4N_aSm(A_0-B_0)}\right\}$$

$$|V_{ab}|^2 = \frac{E_q|q|^2\hbar}{4\rho\omega_{q\lambda}}\left\{\frac{[b_s^\gamma(\theta)\sin2\theta]^2}{16N_aSm(A_0+B_0)} + \frac{[b_c^\gamma(\theta)-f_3(\theta)]^2\sin^2\theta}{4N_aSm(A_0-B_0)}\right\}$$

For $\sin\theta \neq 0$,

$$\begin{aligned}N_aSm(A_0+B_0) &= -6b_2m^3P_2^0(\cos\theta)-20b_4m^{10}P_4^0(\cos\theta)-42b_6m^{21}P_6^0(\cos\theta)\\ &\quad -6b_6^6m^{21}\sin^6\theta\cos6\phi(1-6\cot^2\theta)\\ &\quad +(b_2^cm^3)^2\cos^2\theta(6\sin^2\theta-1)/c^c\\ &\quad +(b_2^\gamma m^3)^2\sin^2\theta(3\sin^2\theta-2)/2c^\gamma\\ &\quad +(b_4^\gamma m^{10})^2\sin^6\theta(5\sin^2\theta-4)/4c^\gamma\\ &\quad +(b_2^\gamma m^3)(b_4^\gamma m^{10})\cos6\phi\sin^4\theta(13\sin^2\theta-10)/4c^\gamma\\ &\quad +2h_cm\cos\theta-h_\perp m\cos2\theta/\sin\theta\\ &\equiv f_2(\theta)+2h_cm\cos\theta-h_\perp m\cos2\theta/\sin\theta\end{aligned}$$

$$N_a Sm(A_0-B_0) = -36b_6^6 m^{21} \sin^4\theta\cos 6\phi + (b_2^\varepsilon m^3)^2 \cos^2\theta/c^\varepsilon$$

$$+ (b_2^\gamma m^3)^2 \sin^2\theta/c^\gamma + (b_4^\gamma m^{10})^2 \sin^6\theta/c^\gamma$$

$$+ 5(b_2^\gamma m^3)(b_4^\gamma m^{10})\cos 6\phi \sin^4\theta/2c^\gamma + h_\perp m/\sin\theta$$

$$\equiv f_3(\theta) + h_\perp m/\sin\theta \quad .$$

The latter two expressions serve to define $f_2(\theta)$ and $f_3(\theta)$. The other quantities appearing in these equations are,

$$b_c^\gamma(\theta) = b_2^\gamma m^3 \cos 2\phi - b_4^\gamma m^{10} \sin^2\theta\cos 4\phi$$

$$b_s^\gamma(\theta) = b_2^\gamma m^3 \sin 2\phi - b_4^\gamma m^{10} \sin^2\theta\sin 4\phi$$

$$h_c = N_a g u_B H_c S$$

$$h_\perp = N_a g u_B (H_a \cos\phi + H_b \sin\phi) S \quad .$$

When $\sin\theta = 0$ the expressions for $f_2(\theta)$ and $f_3(\theta)$ are not valid. Both functions must then be replaced by the function f_1 and the equations above become,

$$N_a Sm(A_0 \pm B_0) = -3b_2 m^3 - 10b_4 m^{10} - 21b_6 m^{21} + h_c m = f_1 + h_c m \quad .$$

REFERENCES

Akhiezer, A. I., Bar'yakhtar, V. G. and Kaganov, M. I., 1961, Soviet Physics Uspekhi 3, 567.

Alstad, J. and Legvold, S., 1964, J. Appl. Phys. 35, 1752.

Atzmony, U., Dariel, M. P., Bauminger, E. R., Lebenbaum, D., Nowik, I. and Ofer, S., 1972, Phys. Rev. Lett. 28, 244.

Atzmony, U., Dariel, M. P., Bauminger, E. R., Lebenbaum, D., Nowik, I. and Ofer, S., 1973, Accepted for Publication in Phys. Rev. B.

Bagguley, D. M. S. and Liesegang, J., 1967, Proc. Roy. Soc. (London) A300, 497.

Bargouth, M. O. and Will, G., 1971, J. de Phys. 32, C1-675.

Becker, R. and Döring, W., 1939, Ferromagnetismus (Berlin, Springer-Verlag).

Bozorth, R. M., 1951, Ferromagnetism (New York, Van Nostrand).

Bozorth, R. M., Clark, A. E. and Gambino, R. J., 1968, Proc. Eleventh Intern. Conf. on Low Temp. Physics, St. Andrews, 1106.

Bozorth, R. M., Clark, A. E. and Van Vleck, J. H., 1972, Intern. J. Magnetism 2, 19.

Brooks, M. S. S. and Goodings, D. A., 1968, J. Phys. C., Ser. 2, 1, 1279.

Brooks, M. S. S., 1970, Phys. Rev. B 1, 2257.

Brooks, M. S. S., 1972, J. Phys. F: Metal Phys. 2, 978.

Brown, W. F., Jr., 1964, Proc. Intern. Conf. on Magnetism, Nottingham, 681.

Brown, W. F., Jr., 1966, Magnetoelastic Interactions (New York, Springer-Verlag).

Buckmaster, H. A., 1962, Can. J. Phys. 40, 1670.

Burzo, E., 1971, Z. Angew. Phys. 32, 127.

Buschow, K. H. J. and van Stapele, R. P., 1970, J. Appl. Phys. 41, 4066.

Callen, E. and Callen, H. B., 1965, Phys. Rev. 139, A455.

Callen, H. B. and Callen, E., 1966, J. Phys. Chem. Solids 27, 1271.

Chow, H. and Keffer, F., 1973, Phys. Rev. B 7, 2028.

Clark, A. E., DeSavage, B. F. and Bozorth, R., 1965, Phys. Rev. 138, A216.

Clark, A. E. and Belson, H. S., 1972a, Phys. Rev. B 5, 3642.

Clark, A. E. and Belson, H. S., 1972b, Trans. IEEE, MAG-8, 477.

Clark, A. E., Belson, H. S. and Tamagawa, N., 1972, Phys. Lett. 42A, 160.

Cooper, B. R., 1968a, Phys. Rev. 169, 281.

Cooper, B. R., 1968b, Sol. St. Phys., edited by F. Seitz and D. Turnbull (Academic Press, New York) 21, 393.

Du Plessis, P. de V., 1968, Phil. Mag. 18, 145.

Elliott, R. J., 1972, Magnetic Properties of Rare Earth Metals (New York, Plenum Press).

Feron, J-L., Hug, G. and Pauthenet, R., 1970, Les Éléments des Terres Rares, CNRS, Paris, 2, 19.

Fisher, E. S. and Dever, D., 1967, Trans. Met. Soc. AIME 239, 48.

Fisher, E. S. and Dever, D., 1968, Proc. of Oak Ridge Conf. on the Rare Earths, 522.

Goodings, D. A. and Southern, B. W., 1971, Can. J. Phys. 49, 1137.

Graham, C. D., Jr., 1966, General Electric Rep. No. 66-C-218.

Graham, C. D., Jr., 1967, J. Appl. Phys. 38, 1375.

Hart, L. W. and Stanford, J. L., 1971, Phys. Rev. Lett. 27, 676.

Holstein, T. D. and Primakoff, H., 1940, Phys. Rev. 58, 1098.

Jensen, J., 1971, Intern. J. Magnetism 1, 271.

Keffer, F., 1966, In Handbuch der Physik, Vol. XVIII/2, edited by S. Flügge (Berlin, Springer-Verlag).

Kittel, C., 1958, Phys. Rev. 110, 836.

Kittel, C. and Van Vleck, J. H., 1960, Phys. Rev. 118, 1231.

Kittel, C., 1963, Quantum Theory of Solids, Chapter 2 (John Wiley and Sons, Inc.).

Kittel, C., 1968, Sol. St. Phys., edited by F. Seitz and D. Turnbull (Academic Press, New York) 21, 1.

Koon, N. C., Schindler, A. I. and Carter, F. L., 1971, Phys. Lett. 37A, 413.

LeCraw, R. C. and Kasuya, T., 1963, Phys. Rev. 130, 50.

Levinson, L. M. and Shtrikman, S. J., 1971, Phys. Chem. Solids 32, 981.

Long, M., Wazzan, A. R. and Stern, R., 1969, Phys. Rev. 178, 775.

Love, A. E. H., 1944, A Treatise on the Mathematical Theory of Elasticity (New York, Dover Publications, Inc.).

Mackintosh, A. R. and Moller, H. B., 1972, In Magnetic Properties of Rare Earth Metals, Chapter 5, edited by R. J. Elliott (New York, Plenum Press).

Marsh, H. and Sievers, A. J., 1969, J. Appl. Phys. 40, 1563.

Mason, W. P., 1954, Phys. Rev. 96, 302.

Melcher, R. L., 1970, Phys. Rev. Lett. 25, 1201.

Melcher, R. L., 1971, Lectures presented at the Enrico Fermi Summer School, Varenna, Italy.

Moller, H. B., Houmann, J. C. and Mackintosh, A. R., 1967, Phys. Rev. Lett. 19, 312.

Moran, T. J. and Lüthi, B., 1970, J. Phys. Chem. Solids 31, 1735.

Murnaghan, F. D., 1951, Finite Deformation of an Elastic Solid (John Wiley and Sons, Inc.).

Nayyar, A. H. and Sherrington, D., 1972, J. Phys. F. 2, 893.

Nielsen, M., Moller, H. B. and Mackintosh, A. R., 1970a, J. Appl. Phys. 41, 1174.

Nielsen, M., Moller, H. B., Lindgard, P. A. and Mackintosh, A. R., 1970b, Phys. Rev. Lett. 25, 1451.

Palmer, S. B. and Lee, E. W., 1972, Proc. Roy. Soc. A327, 519.

Pollina, R. J. and Lüthi, B., 1969, Phys. Rev. 177, 841.

Rhyne, J. J. and Legvold, S., 1965a, Phys. Rev. 138, A507.

Rhyne, J. J. and Legvold, S., 1965b, Phys. Rev. 140, A2143.

Rhyne, J. J., Legvold, S. and Rodine, E. T., 1967, Phys. Rev. 154, 266.

Rhyne, J. J. and Clark, A. E., 1967, J. Appl. Phys. 38, 1379.

Rhyne, J. J., Foner, S., McNiff, E. J., Jr., and Doclo, R., 1968, J. Appl. Phys. 39, 892.

Rosen, M., 1967, Phys. Rev. Lett. 19, 695.

Rosen, M. and Klimker, H., 1970, Phys. Rev. B 1, 3748.

Rossol, F. C. and Jones, R. V., 1966, J. Appl. Phys. 37, 1227.

Schlömann, E., 1960, J. Appl. Phys. 31, 1647.

Slanicka, M. I., Taylor, K. N. R. and Primavesi, G. J., 1971, J. Phys. F: Metal Phys. 1, 679.

Smit, J. and Belgers, H. G., 1955, Philips Res. Repts. 10, 113.

Southern, B. W. and Goodings, D. A., 1973, Phys. Rev. B 7, 534.

Southern, B. W., 1973, Accepted for Publication in Can. J. Phys.

Taylor, K. N. R. and Darby, M. I., 1972, Physics of Rare Earth Solids (Chapman and Hall Ltd.).

Tiersten, H. F., 1964, J. Math. Phys. 5, 1298.

Toupin, R. A., 1956, J. Ratl. Mech. Anal. 5, 849.

Truesdell, C. and Toupin, R. A., 1960, In Handbuch der Physik, Vol. III, edited by S. Flügge (Berlin, Springer-Verlag).

Turov, E. A. and Shavrov, V. G., 1965, Soviet Phys. - Solid State 7, 166.

Vigren, D. T. and Liu, S. H., 1972, Phys. Rev. B 5, 2719.

Wagner, T. K. and Stanford, J. L., 1969, Phys. Rev. 184, 505.

Wagner, T. K. and Stanford, J. L., 1972, Phys. Rev. B 5, 1876.

Wortis, M., 1963, Ph.D. Thesis, Harvard University.