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Effect of Uniaxial Stress on the Helimagnetic-Ferromagnetic
Phase Transition in Single-Crystal Dysprosium

by

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A THESIS SUBMITTED
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE OF
MASTER OF ARTS

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Houston, Texas

May, 1970

ABSTRACT

Effect of Uniaxial Stress on the Helimagnetic-Ferromagnetic Phase Transition in Single-Crystal Dysprosium

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The effect of an applied uniaxial stress on the value of the critical field, at which the helimagnetic-ferromagnetic phase transition occurs, in single-crystal dysprosium has been studied. Measurements were made with stress applied along the hexagonal axis, at temperatures between 85 K and 179 K, and the maximum stress employed was 600 bar. This is comparable to the effective internal stress due to magnetostriction naturally occurring in dysprosium. At all temperatures in the range studied, the critical field increased linearly with increasing stress; the maximum shift in the critical field was approximately 1.5 kOe. This increase in the critical field can be attributed qualitatively to the fact that the application of compressive stress opposes the normal magnetostrictive expansion of the hexagonal axis which accompanies the phase transition. As a result, a larger magnetic field is required in order to overcome the tendency toward helical ordering due to the nature of the exchange energy. A comparison of the experimental results with theoretical predictions based on the usual phenomeno-

logical Hamiltonian permits a more accurate determination of the magnetoelastic contributions to this Hamiltonian than has been previously reported.

To Donna

TABLE OF CONTENTS

	Page
INTRODUCTION	1
THEORY	8
I. The Hamiltonian	15
A. The Anisotropy Hamiltonian	15
B. The Exchange Hamiltonian	17
C. The Magnetoelastic Hamiltonian	19
D. The Elastic Energy	29
E. The Zeeman Hamiltonian	30
II. The Gibbs Free Energy	31
III. The Critical Field	38
APPARATUS AND EXPERIMENTAL PROCEDURE	40
I. Sample Preparation	41
II. Cryogenic System	43
III. Stress Mechanism and Sample Support	46
IV. Detector and Dummy Coils	50
V. Electronic Equipment	51
RESULTS AND DISCUSSION	53
BIBLIOGRAPHY	59
ACKNOWLEDGMENTS	62

INTRODUCTION

Dysprosium is the sixty-sixth element of the periodic table, belonging to the Lanthanide series or rare-earth group. It has a melting point of 1680 K, a boiling point of 2873 K, and specific gravity of 8.536. The metal has a hexagonal close-packed crystal structure. A Dy atom has an electronic configuration of $4f^9 5d^1 6s^2$ plus a xenon core. In the metal, the valence electrons $5d^1 6s^2$ are lost to conduction bands; then at each lattice site there is left a highly localized magnetic moment due to unpaired electrons in the 4f shell. The 4f electrons, which occupy a deep shell, shielded by filled 5s and 5p shells, play a secondary role in the chemical and mechanical properties of dysprosium. On the other hand, they are the origin of the magnetic properties. The conduction electrons enter into the magnetic properties primarily as a means of exchange coupling between localized 4f electrons.

Much of the fundamental interest in dysprosium lies in just this point, that to a very good approximation one can make the distinction between localized magnetic electrons which carry the major part of the bulk magnetic moment and itinerant conduction electrons which contribute little mag-

netic moment. This model successfully explains the various equilibrium magnetic properties observed; for example, the effective paramagnetic moment at high temperatures is nearly identical to that expected for an assembly of free tripositive ions.

There are several interesting features in the magnetic properties of dysprosium metal^(1,2). A Dy crystal becomes magnetically ordered at a Neel point $T_n = 179$ K. At this temperature Wilkinson et al. have established that there is ferromagnetic ordering in each basal plane of the hcp lattice, and that the magnetic moment in successive planes is turned through an initial turn angle $\varphi_n = 43^\circ$, forming a helimagnetic (HM) structure. Further, the turn angle decreases continuously with temperature, reaching a value of $\varphi_c = 26.5^\circ$ at the Curie point $T_c = 85$ K. At this temperature the turn angle decreases abruptly to zero in a first-order phase transition, and a ferromagnetic (FM) structure is formed.

The spontaneous magnetization always lies in the basal plane of the hcp crystal. The magnetic moment of the metal is linearly dependent on the value of an external magnetic field when it is applied parallel to the "c" axis of the

FERROMAGNETIC

HELMAGNETIC

MAGNETIC STRUCTURES
OF DYSPROSIUM

FIGURE I

Figure 2

Dysprosium Magnetization

Figure 3

crystal at all temperatures for fields up to approximately 165 kOe; that is, the crystal is very hard magnetically in the direction of the "c" axis⁽³⁾. At low temperatures the direction of easy magnetization is along the six equivalent $[11\bar{2}0]$ directions (the directions along the line joining nearest neighbors in the basal plane of the hexagonal lattice, or "a" directions), but above 110 K the magnetization is isotropic in the basal plane⁽¹⁾. In the HM temperature range the Zeeman energy due to quite moderate fields applied in the basal plane is sufficient to cause the transition to the FM phase. The critical field H_c necessary to cause this phase transition increases almost linearly with temperature to a maximum value of approximately 12 kOe near T_n and then decreases rapidly to zero at T_n .

In dysprosium the onset of ferromagnetism at 85 K is accompanied by a transition from a hexagonal to an orthorhombic structure⁽⁴⁾. Figure 5 shows that when the helimagnetic structure is converted to a ferromagnetic structure by the application of a field in the basal plane above 85 K, the accompanying magnetostriction produces a similar orthorhombic distortion⁽⁵⁾. For no externally applied stresses the "c" axis expands, and in the basal plane there is an expansion parallel to the magnetic field and a contraction

Figure 4

FIGURE 5

perpendicular to the field. This giant magnetostrictive effect (typical strains of 10^{-3} - 10^{-2}) is 2-3 orders of magnitude larger than for any transition metal⁽⁶⁾.

Enz⁽⁷⁾ first pointed out that the occurrence of a stable helical spin structure for Dy could be explained as the result of competing exchange interactions: a large positive interaction between nearest neighbors to align each basal plane ferromagnetically, a positive interaction between second-nearest neighbors, and a negative interaction between third-nearest neighbors. The transition from HM to FM arrangement at T_c is governed by the competition between the exchange interaction for the undistorted hcp lattice favoring HM arrangement on the one hand, and the magnetostriction or the hexagonal anisotropy effects (of the undistorted lattice) favoring FM alignment on the other hand. The reason that the transition occurs is that the temperature dependence of these different contributions to the free energy differ sharply. The magnetostriction and planar anisotropy terms are larger at low temperature but fall off more rapidly as it increases so that at the high temperature end of the HM regime, exchange effects for the undistorted lattice dominate⁽⁸⁾. The planar anisotropy energy falls off very rapidly with increasing temperature near T_c , becoming

negligible for temperatures above 110 K; thus, it acts as the driving force for the HM to FM transition, although the magnetoelastic energy is larger. For the field-induced transition at H_c , the Zeeman energy plays the same role as the planar anisotropy energy near T_c in aiding the magnetoelastic energy to bring about the transition to FM alignment of the moments.

The HM-FM transition is a first-order thermodynamic phase transition, as indicated by the sudden changes of the magnetization⁽¹⁾, crystallographic parameters⁽⁴⁾, the shape of the specific heat peaks⁽⁹⁾, and resistivity⁽¹⁰⁾. First-order transition is used here in its usual thermodynamic sense, i.e. the system under consideration possesses an equilibrium free energy that is a function of temperature and pressure and the first derivative of that free energy function with either temperature or pressure exhibits a discontinuity at a critical value of temperature or pressure. Thus the entropy and/or the volume of the system has a discontinuity. This is not the situation for a second-order transition in which case it is the second derivatives that suffer discontinuities, i.e. the thermal expansion, specific heat, and compressibility⁽¹¹⁾.

The dependence of magnetism upon interatomic distance provides information which should lead to a deeper understanding of this remarkable phenomenon. One of the simplest and most direct ways of obtaining experimental data related to this question for ferromagnetism and antiferromagnetism is to measure the stress dependence of magnetic transitions. Several authors have investigated the pressure dependence of the transition temperatures T_n and T_c for dysprosium⁽¹²⁻¹⁸⁾. A marked discrepancy exists among the values reported for the pressure dependence of the Curie temperature T_c . The sharp temperature dependence of the magnetoelastic and planar anisotropy energies causes the shift of T_c with pressure to be a small effect. On the other hand, the Zeeman energy is a linear function of an applied field, thus one would expect the shift of H_c due to an applied stress to be a much larger effect.

The effect of an applied uniaxial stress on the critical field H_c was measured in this experiment. The sharp increase in magnetization at H_c produces a susceptibility (dM/dH) maximum at that point. The stress was applied along the "c" axis of a Dy single crystal, and the resulting shift in the susceptibility maximum was measured. The maximum stress employed was 600 bar. This is comparable to the

the effective internal stress due to magnetostriction at the transition. At all temperatures in the HM regime the critical field increased linearly with increasing stress; the maximum shift in the critical field was approximately 1.5 kOe. This increase in the critical field can be attributed qualitatively to the fact that the application of compressive stress opposes the normal magnetostrictive expansion of the "c" axis which accompanies the phase transition. As a result, a larger magnetic field is required in order to overcome the tendency toward helical ordering due to the nature of the exchange energy. A comparison of the experimental results with theoretical predictions based on the usual phenomenological Hamiltonian permits a more accurate determination of the magnetoelastic contributions to this Hamiltonian than has been previously reported.

THEORY

The bulk magnetic properties of single-crystal dysprosium can be attributed to the hcp lattice of highly localized magnetic moments associated with unfilled 4f shells. The conduction electrons play a key role in the indirect exchange coupling between Dy ions and in their effect on the crystalline field. Band calculations^(19,20) indicate, however, that the conduction bands and Fermi surface probably have the mixed s-d character of the type found in transition and noble metals, with large departure from free-electron-like behavior. The lack of experimental information about the Fermi surface thus makes meaningful calculations of the interactions involving the conduction electrons impossible. Therefore, a phenomenological treatment of the strongly coupled 4f shell electrons is often used in theoretical formulations of the magnetic properties of the rare earths.

A general spin Hamiltonian for a magnetic moment system is constructed based upon physical considerations of the contributions to the magnetic energy and each term is required to have the symmetry of the hcp lattice. This procedure yields a set of approximately temperature-independent phenomenological constants capable of describing the observed

magnetic properties of the system. The molecular field approximation may be used to obtain the energy of the system for the helimagnetic (HM) and ferromagnetic (FM) structures. The molecular field hypothesis states that the effective field acting on a moment is proportional to the magnetization for no applied field; thus, the magnetic Hamiltonian corresponding to exchange interactions is proportional to $\vec{J}_i \cdot \sum_{j \neq i} \langle \vec{J}_j \rangle$. The molecular field hypothesis is therefore equivalent to the assumption that $\langle \vec{J}_i \cdot \vec{J}_j \rangle_{i \neq j} = \langle \vec{J}_i \rangle \cdot \langle \vec{J}_j \rangle$.

For a system in equilibrium with a reservoir at constant temperature and stress, the appropriate thermodynamic quantity to consider is the Gibbs free energy G . The equilibrium condition is that the Gibbs free energy G of the system is a minimum. The system of magnetic moments can exist in either the HM phase or the FM phase or a mixture of the two. The critical field H_c for a given temperature and stress corresponds to the magnetic field at which the two phases can coexist in equilibrium with each other. If $G^{HM}(H_c)$ is the helimagnetic Gibbs free energy per ion and $G^{FM}(H_c)$ is the ferromagnetic Gibbs free energy per ion, the total Gibbs free energy for the system is given by

$$G(H_c) = N_{HM} G^{HM}(H_c) + N_{FM} G^{FM}(H_c) , \quad (1)$$

where N_{HM} and N_{FM} are the number of ions in the HM and FM phases respectively. The total number of ions $N = N_{\text{HM}} + N_{\text{FM}}$ is constant, thus $dN = dN_{\text{HM}} + dN_{\text{FM}} = 0$. Therefore the requirement that $dG = 0$ for equilibrium becomes

$$[G^{\text{HM}}(H_c) - G^{\text{FM}}(H_c)] dN_{\text{HM}} = 0 ,$$

or
$$G^{\text{HM}}(H_c) = G^{\text{FM}}(H_c) . \quad (2)$$

Using the molecular field hypothesis and the magnetic Hamiltonian, the Gibbs free energy function is constructed for each phase (HM and FM) and minimized with respect to the strains. The free energies for the two phases are then set equal, and the critical field H_c is found as a function of applied stress.

Behrendt et al.⁽¹⁾ found the paramagnetic susceptibility of Dy to be nearly identical to that of an assembly of tri-positive free ions. The susceptibility of such an assembly of ions is given by the Curie-Weiss law, $\chi = \frac{C}{T-\theta}$, where $C = Ng^2\beta^2J(J+1) / (3kV)$ and θ is the paramagnetic Curie temperature. The Curie-Weiss Law is based on the molecular field hypothesis, using Boltzman statistics, for N similar ions (in a volume V) restricted to the lowest J multiplet, where J is given by Hund's rule, g is Lande's g -factor, β

is the Bohr magneton, k is the Boltzmann constant, and T is the temperature. Hund's rule predicts $J = 15/2$ for the nine 4f electrons in Dy. The saturation moment exceeds, slightly, the value predicted from the Hund's rule ground state, $g\beta J^{(21)}$. The excess is usually attributed to polarization of the conduction electrons. The close agreement of the paramagnetic susceptibility with the Curie-Weiss law indicates that the molecular field hypothesis and the assumption of highly localized moments are valid approximations.

The success of the free-ion susceptibility behavior can be explained qualitatively in the following manner. For a particular ion, the complete 5s and 5p shells, plus the conduction electrons, shield the 4f electrons from the crystalline field due to the remaining ions. Thus it is plausible to assume that the total orbital angular momentum of the 4f electrons is not quenched by the crystalline field. With this assumption we expect the ionic ground state in the metal to be the same as the Hund's rule ground state of a free ion. The next excited J multiplet is higher in energy by more than $10^3 K$ so that it plays no role in thermal properties⁽²²⁾. For this reason it is always sufficient to regard the ions as being confined to the lowest J multiplet.

In view of the calculations to be presented, the zero-field reduced spontaneous magnetization $\sigma(T)$ is an important parameter. In the FM regime $\sigma(T)$ is unambiguously defined as $\sigma(T) \equiv \frac{M(T)}{M(0)}$, where $M(T)$ is the spontaneous magnetization at the temperature T . In the HM regime, $\sigma(T)$ is defined by the spontaneous magnetization of a layer of ions, $M_L(T)$, in the basal plane; $\sigma(T) \equiv \frac{M_L(T)}{M_L(0)}$. $M_L(T)$ is obtained by extrapolating the high field magnetization curve (where there is FM alignment) down to zero field. Since

$$\frac{M(T)}{M(0)} = \frac{ng\beta\langle M_J \rangle}{ng\beta J},$$

a more fundamental definition is $\sigma(T) \equiv \frac{\langle M_J \rangle}{J}$.

Thus, using the molecular field approximation for terms such as $\vec{J}_i \cdot \vec{J}_j$, we have

$$\langle \vec{J} \cdot \vec{J} \rangle_{i \neq j} = \langle \vec{J}_i \rangle \cdot \langle \vec{J}_j \rangle = J^2 \sigma(T)^2 \cos \alpha, \quad (3)$$

where α is the angle between $\langle \vec{J}_i \rangle$ and $\langle \vec{J}_j \rangle$.

The temperature dependence of σ can be calculated theoretically using the molecular field hypothesis, Boltzmann statistics, and restricting J to be that of the ground-state multiplet. The simplest approximation is to ignore the HM phase, that is assume FM order for temperatures up to the Neel point. This simple approximation for

REDUCED SPONTANEOUS MAGNETIZATION

FIGURE 6

σ can be expressed as⁽²³⁾

$$\sigma(T) = B_J \left(\frac{3J}{J+1} \frac{T_N}{T} \sigma(T) \right) , \quad (4)$$

where $B_J(x)$ is the Brillouin function,

$$B_J(x) = \left(\frac{2J+1}{2J} \right) \coth \left(\frac{2J+1}{2J} x \right) - \frac{1}{2J} \coth \frac{x}{2J} . \quad (5)$$

This transcendental equation for σ can be solved graphically, and the result is compared to the experimental values in Figure 6 . The agreement is not excellent, but it is reasonable since the HM phase is not considered.

It is obvious from the magnetization curves of Figure 3 , that the HM structure is distorted by an applied field in the basal plane before the critical field H_c is reached. Theoretical studies of the magnetization process of a helical spin structure were first made by Herpin, Meriel, and Villain⁽²⁴⁾, and by Enz⁽⁷⁾. For small fields the theory predicts that there is a slight distortion of the helix with a small net moment along H. At a critical field H_c there is an abrupt transition to a fanlike structure with a large net moment along H. As the field increases still further, the angular amplitude decreases continuously until complete ferromagnetic alignment is achieved. For sufficiently large

planar anisotropy, Cooper and Elliott⁽²⁵⁾ have shown that the fan phase disappears, and the transition at H_c is directly to the ferromagnet. Presumably magnetostriction can have the same effect. For Dy the planar anisotropy is large enough so that there probably is no fan phase, and the transition is directly from helix to ferromagnet⁽⁸⁾. In the calculations to be presented, the magnetization process is assumed to be directly from an undistorted helimagnetic structure to a ferromagnetic structure. This simplified approach ignores the field induced magnetization and the corresponding Zeeman energy for fields less than H_c (Figure 7).

MAGNETIZATION MODEL

FIGURE 7

I. The Hamiltonian

The variations in the magnetic properties of a dysprosium single crystal can be considered the natural consequence of a Hamiltonian for the 4f localized moment system:

$$\mathcal{H} = \mathcal{H}_a + \mathcal{H}_{ex} + \mathcal{H}_{me} + \mathcal{H}_e + \mathcal{H}_z . \quad (6)$$

The Anisotropy Hamiltonian

The first term in the Hamiltonian represents the anisotropy energy of the unstrained lattice. There may be several kinds of anisotropy energy with different atomic origins in dysprosium. One of the most important terms is the one due to the crystalline field acting on the distribution of currents possessed by the 4f electrons⁽²⁶⁾. In Dy the crystalline field is weak due to the shielding of the 4f shell, but the large orbital angular momentum interacts strongly with this field. The theory of such fields has been extensively developed in ionic crystals. The potential energy of a single electron at (r, θ, φ) can be expanded in spherical harmonics as

$$V = \sum_{n,m} V_n^m r^n Y_n^m(\theta, \varphi) . \quad (7)$$

For hexagonal symmetry and considering the lowest order terms, this becomes⁽²⁷⁾

$$V = V_2^0 r^2 Y_2^0(\theta, \varphi) + V_4^0 r^4 Y_4^0(\theta, \varphi) + V_6^0 r^6 Y_6^0(\theta, \varphi) \\ + V_6^6 r^6 [Y_6^6(\theta, \varphi) + Y_6^{-6}(\theta, \varphi)] \quad .$$

Within the ground-state J manifold, the total anisotropy Hamiltonian due to the crystalline field may be written in terms of operator equivalents of spherical harmonics⁽²⁸⁾ as

$$\mathcal{H}_a = \sum_i \left\{ P_2 Y_2^0(\vec{J}_i) + P_4 Y_4^0(\vec{J}_i) + P_6 Y_6^0(\vec{J}_i) \right. \\ \left. + P_6^6 [Y_6^6(\vec{J}_i) + Y_6^{-6}(\vec{J}_i)] \right\} \quad . \quad (8)$$

This can be written in the simpler form

$$\mathcal{H}_a = \sum_i \left\{ K_2 J_{iz}^2 + K_4 J_{iz}^4 + K_6 J_{iz}^6 - K_6^6 [J_{i+}^6 + J_{i-}^6] \right\} \quad (9)$$

where the constants are taken to be positive, as found by Miwa and Yosida⁽²⁹⁾. With this convention for K_6^6 , $\hat{x}$ is parallel to the "a" axis of the crystal, and $\hat{z}$ is parallel to the "c" axis.

The magnetic dipolar interactions between ions in Dy contribute to the anisotropy energy, but under the assumption of the molecular field approximation it is found that they are isotropic in the basal plane of the crystal and produce

very small anisotropy energy in the "c" direction⁽³⁰⁾.

Another possibility has been suggested for the source of the large magnetic anisotropy in Dy. This is anisotropic exchange and can arise because of the large orbital contribution to the magnetic moment. Several authors^(31,32) have explored the original suggestions of Kaplan and Lyons⁽³³⁾ along these lines.

Magnetization measurements⁽¹⁾ indicate that the axial terms of the anisotropy energy (e.g. the first three terms of Equation 9) are quite large, restricting the moments to lie entirely in the basal plane. These measurements also indicate that the basal plane anisotropy terms (e.g. the last term in Equation 9) are negligible for temperatures greater than 110 K. For a first approximation the axial anisotropy can be included implicitly by requiring $\langle J_{iz} \rangle = 0$, and the basal plane anisotropy can be ignored; thus the anisotropy Hamiltonian of the unstrained lattice is not explicitly included in the general Hamiltonian.

The Exchange Hamiltonian

The second term in the Hamiltonian represents the exchange interaction of the unstrained lattice. As it was pointed out in the earliest papers describing a helical magnetic moment structure⁽³⁴⁻³⁸⁾, the existence of a stable

helimagnetic structure depends on the presence of long-range competing exchange interactions. Direct exchange between ions in Dy is small since the 4f orbitals have negligible overlap; but, it is generally accepted that an indirect exchange interaction via the conduction electrons, as proposed by Ruderman and Kittel⁽³⁹⁾ and expanded by Kasuya⁽⁴⁰⁾ and by Yosida⁽⁴¹⁾, is quite large in Dy. This Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction takes the Heisenberg form

$$\mathcal{H}_{\text{ex}} = - \sum_{i \neq j} I_{ij} \vec{J}_i \cdot \vec{J}_j \quad . \quad (10)$$

1. Helimagnetic Phase

The spin components for the HM structure can be described by

$$\langle J_{iz} \rangle = 0 \quad , \quad \langle J_{ix} \rangle = J\sigma \cos\varphi_i \quad , \quad \text{and} \quad \langle J_{iy} \rangle = J\sigma \sin\varphi_i \quad , \quad (11)$$

where $\hat{z} \parallel c$ and $\hat{x} \perp \hat{y}$ in the basal plane. Using the molecular field approximation,

$$\langle J_i \cdot J_j \rangle = J^2 \sigma^2 [\cos\varphi_i \cos\varphi_j + \sin\varphi_i \sin\varphi_j] = J^2 \sigma^2 \cos(\varphi_i - \varphi_j) \quad . \quad (12)$$

For simplicity we assume nearest, next-nearest, and third-nearest neighbor interactions. Let I'_0 be the exchange interaction between the six nearest neighbors (in the basal plane),

EXCHANGE INTERACTIONS

FIGURE 9

I'_1 be the exchange interaction between the six next-nearest neighbors (in the adjoining two planes), and I'_2 be the exchange interaction between the two third-nearest neighbors (see Figure 9). The total exchange energy then becomes

$$\langle \mathcal{H}_{\text{ex}} \rangle = -NJ^2\sigma^2 [6I'_0 + 6I'_1\cos\varphi + 2I'_2\cos 2\varphi] , \quad (13)$$

where φ is the HM turn angle between adjacent basal planes.

By defining $I_0 = 6J^2I'_0$, $I_1 = 6J^2I'_1$, and $I_2 = 2J^2I'_2$ the exchange energy per ion is given by

$$E_{\text{ex}}^{\text{HM}} = -\sigma^2 [I_0 + I_1\cos\varphi + I_2\cos 2\varphi] . \quad (14)$$

2. Ferromagnetic Phase

For an applied field $\vec{H} \parallel \hat{x}$ the FM exchange energy per ion is given by Equation 14 with $\varphi = 0$.

$$E_{\text{ex}}^{\text{FM}} = -\sigma^2 [I_0 + I_1 + I_2] . \quad (15)$$

The Magnetoelastic Hamiltonian

The third term in the Hamiltonian represents the magnetoelastic energy. In a ferro- or antiferromagnetic crystal the moment system is coupled to the ionic displacements by the inter-ionic distance dependence of the moment-interaction energies, such as anisotropy, exchange, dipolar, pseudo-dipolar, and exchange energies. Callen and Callen⁽⁴²⁾

have formulated a general phenomenological treatment for the magnetoelastic energy that is independent of its atomic origins. The procedure is as follows. For small strains, consider only terms linear in the strain components. Also, for a first approximation, consider angular momentum components of order two. Linear terms in angular momentum components are excluded because of antisymmetry with respect to time reversal. A general Hamiltonian is constructed using these terms, and symmetry requirements are used to simplify this Hamiltonian.

1. The Single-ion Magnetoelastic Hamiltonian

The overriding contribution to the large single-ion magnetoelastic coefficients is from the change in electrostatic energy between conduction and 4f electrons⁽⁴³⁾. Consider the anisotropic 4f charge cloud, with its symmetry axis along the magnetization. The change in crystal field accompanying distortion causes a change in electrostatic energy of the 4f electrons, which change depends upon magnetization direction. Suppose an oblate charge cloud in the yz plane has its orbital angular momentum along x in the basal plane. Neighboring + ions along y are attracted inward and + ions along x tend to move away (no volume strain) to minimize the electrostatic energy between

4f electrons and neighboring cores. Conduction-electron redistribution accompanies the lattice distortion⁽⁴²⁾.

Using the theory of Callen and Callen⁽⁴²⁾, the general form for the single-ion contribution to the magnetoelastic Hamiltonian is given by

$$H_{me}^I = \sum_{f=1}^N \sum_{i,j,k,l=1}^3 B_{ijkl} \epsilon_{ij}^f \epsilon_{fk}^f \epsilon_{fl}^f, \quad (16)$$

where f labels the ion sites. The classical polynomial basis functions for the chemical point group D_{3h} of the hcp lattice are

$$(x^2+y^2+z^2)_\alpha, \quad \frac{\sqrt{3}}{2} (z^2 - \frac{1}{3} r^2)_\alpha, \quad [yz, xz]_\epsilon, \quad \text{and} \\ [(x^2-y^2)/2, xy]_\gamma.$$

The strain components are isomorphic to these polynomials; hence the symmetry strains are

$$\begin{aligned} \epsilon^{\alpha,1} &= \epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz}, \\ \epsilon^{\alpha,2} &= \frac{\sqrt{3}}{2} \left(\frac{2}{3} \epsilon_{zz} - \frac{1}{3} \epsilon_{xx} - \frac{1}{3} \epsilon_{yy} \right), \\ \epsilon_1^\epsilon &= \epsilon_{yz}, \quad \epsilon_2^\epsilon = \epsilon_{xz}, \\ \epsilon_1^\gamma &= \frac{1}{2} (\epsilon_{xx} - \epsilon_{yy}), \quad \text{and} \quad \epsilon_2^\gamma = \epsilon_{xy}. \end{aligned} \quad (17)$$

(Notation: $\epsilon_i^{\Gamma,j}$; Γ = an irreducible representation,

$i = 1 \rightarrow n$ where n is the dimensionality of Γ , $j = 1 \rightarrow m$

where m = number of basis functions belong to Γ .) Similarly, there are six symmetry angular momentum operators of second degree that are fully analogous to the strain components, if we remember that angular momentum operators do not in general commute. For example, $\epsilon_{xy} \rightarrow \frac{1}{2} (J_x J_y + J_y J_x)$. Thus the symmetry angular momentum operators are

$$\begin{aligned} J_i^{\alpha,1} &= J_{ix}^2 + J_{iy}^2 + J_{iz}^2 = \vec{J}_i \cdot \vec{J}_i = J(J+1) \\ J_i^{\alpha,2} &= \frac{\sqrt{3}}{2} [J_{iz}^2 - \frac{1}{3} J(J+1)] , \\ J_{i1}^{\epsilon} &= \frac{1}{2} (J_{iy} J_{iz} + J_{iz} J_{iy}) , \quad J_{i2}^{\epsilon} = \frac{1}{2} (J_{ix} J_{iz} + J_{iz} J_{ix}) , \\ J_{i1}^{\gamma} &= \frac{1}{2} (J_{ix}^2 - J_{iy}^2) , \quad \text{and} \quad J_{i2}^{\gamma} = \frac{1}{2} (J_{ix} J_{iy} + J_{iy} J_{ix}) . \end{aligned} \quad (18)$$

Using the symmetry strains and angular momentum operators the single-ion magnetoelastic Hamiltonian can be expressed as

$$\mathcal{H}_{me}^I = - \sum_f \sum_{\Gamma} \sum_{j,j'} B_{jj'i}^{\Gamma} \sum_i \epsilon_i^{\Gamma,j} J_{fi}^{\Gamma,j'} . \quad (19)$$

The spin operators are linear combinations of spherical tensor operators Y_{ℓ}^m for $\ell = 0, 2$. Hence

$$\langle J_{fi}^{\Gamma,j} \rangle = \sum_m a_{i\Gamma j}^m \langle Y_{\ell}^m \rangle . \quad (20)$$

We re-express the spherical tensor operators in a rotated

(ξ, η, τ) coordinate system, with the τ axis along the direction of magnetization $\vec{\alpha}_f$ for the ion labeled f . Let the spherical tensor operators in this new coordinate system be distinguished by a tilde. Then

$$Y_\ell^m = \sum_{m'} \langle \tilde{Y}_\ell^{m'}(f) | Y_\ell^m \rangle \tilde{Y}_\ell^{m'}(f) \quad . \quad (21)$$

If we assume the unperturbed Hamiltonian for the ion f to have azimuthal symmetry about $\vec{\alpha}_f$, only the $m' = 0$ term remains after taking the average. Thus

$$\langle Y_\ell^m \rangle = \langle \tilde{Y}_\ell^0(f) | Y_\ell^m \rangle \langle \tilde{Y}_\ell^0(f) \rangle \quad . \quad (22)$$

Furthermore,

$$\langle \tilde{Y}_\ell^0(f) | Y_\ell^m \rangle = y_\ell^m(\vec{\alpha}_f) \quad (23)$$

where $y_\ell^m(\vec{\alpha})$ is the spherical harmonic whose arguments θ_f and φ_f are those of the magnetization direction $\vec{\alpha}_f$ relative to the crystal axes. Thus

$$\langle J_{fi}^{\Gamma, j} \rangle = \langle \tilde{Y}_\ell^0(f) \rangle \sum_m a_{i\Gamma j}^m y_\ell^m(\vec{\alpha}_f) \quad , \quad (24)$$

or

$$\langle J_{fi}^{\Gamma, j} \rangle = \langle \tilde{Y}_\ell^0(f) \rangle K_i^{\Gamma, j}(\vec{\alpha}_f) \quad , \quad (25)$$

where $K_i^{\Gamma, j}(\vec{\alpha}_f)$ is the same function of α_x , α_y and α_z as

$J_i^{\Gamma,j}(f)$ is of J_{fx} , J_{fy} and J_{fz} . That is

$$\begin{aligned}
 K_f^{\alpha,1} &= \alpha_{fx}^2 + \alpha_{fy}^2 + \alpha_{fz}^2 = 1, \quad (\ell = 0 \text{ term}) \\
 K_f^{\alpha,2} &= \frac{\sqrt{3}}{2} [\alpha_{fz}^2 - \frac{1}{3}] , \\
 K_{f1}^{\epsilon} &= \alpha_{fy}\alpha_{fz}, \quad K_{fz}^{\epsilon} = \alpha_{fx}\alpha_{fz} \\
 K_{f1}^{\gamma} &= \frac{1}{2} (\alpha_{fx}^2 - \alpha_{fy}^2), \quad \text{and} \quad K_{fz}^{\gamma} = \alpha_x\alpha_y . \quad (26)
 \end{aligned}$$

Since calculation of $\langle \tilde{Y}_\ell^0(f) \rangle$ is tedious, the results obtained by Callen and Callen⁽⁴²⁾ will only be quoted. They found that $\langle \tilde{Y}_\ell^0(f) \rangle$ can be expressed as a function of the reduced magnetization $\sigma(T,H)$ and that the results for moderate spin rapidly converge toward the results for infinite spin. For infinite spin we have

$$\frac{\langle \tilde{Y}_\ell^0(f;T,H) \rangle}{\langle \tilde{Y}_\ell^0(f;0,0) \rangle} = \frac{I_{\ell+1/2}(x)}{I_{1/2}(x)} \equiv \hat{I}_{\ell+1/2}(x) , \quad (27)$$

where $I_{\ell+1/2}(x)$ is the hyperbolic Bessel function of order $\ell+1/2$. The argument x is to be considered as a parameter to be expressed in terms of the reduced magnetization by inversion of the relationship

$$\sigma(T,H) = \frac{\langle \tilde{Y}_1^0(f;T,H) \rangle}{\langle \tilde{Y}_1^0(f;0,0) \rangle} = \coth x - \frac{1}{x} = \mathcal{L}(x) , \quad (28)$$

where $\mathcal{L}(x)$ is the familiar Langevin function. We further assume the reduced magnetization to be field independent for an approximation; and since all ions in the crystal are identical, we expect $\langle \tilde{Y}_\ell^0(f) \rangle$ to be independent of f (i.e. the same for all ions).

Using these results, and the conditions $\alpha_{iz} = 0$, $\alpha_{ix} = \cos\varphi_i$ and $\alpha_{iy} = \sin\varphi_i$, the single-ion magnetoelastic energy can be expressed as

$$\begin{aligned}
 \langle \mathcal{H}_{me}^I \rangle = & NB_1 \epsilon_3 - NB_2 (\epsilon_1 + \epsilon_2) - NB_3 \hat{I}_{5/2}(\mathcal{L}^{-1}(\sigma)) \epsilon_3 \\
 & - NB_4 \hat{I}_{5/2}(\mathcal{L}^{-1}(\sigma)) (\epsilon_1 + \epsilon_2) \\
 & - B_5 \hat{I}_{5/2}(\mathcal{L}^{-1}(\sigma)) (\epsilon_1 - \epsilon_2) \sum_i \cos 2\varphi_i \\
 & - 2B_5 I_{5/2}(\mathcal{L}^{-1}(\sigma)) \epsilon_6 \sum_i \sin 2\varphi_i \quad . \quad (29)
 \end{aligned}$$

In Equation 29 the Voigt notation was used, that is

$$\begin{aligned}
 \epsilon_{xx} &= \epsilon_1, \quad \epsilon_{yy} = \epsilon_2, \quad \epsilon_{zz} = \epsilon_3, \quad \epsilon_{yz} = \epsilon_4, \quad \epsilon_{xz} = \epsilon_5, \\
 \text{and } \epsilon_{xy} &= \epsilon_6.
 \end{aligned}$$

a. Helimagnetic Phase

For the HM phase $\sum_i \cos 2\varphi_i = 0$ and $\sum_i \sin 2\varphi_i = 0$, thus the single-ion magnetoelastic energy per ion is given by

$$E_{me}^I(\text{FM}) = - \left\{ B_1 \epsilon_3 + B_2 (\epsilon_1 + \epsilon_2) + B_3 \hat{I}_{5/2}(\mathcal{L}^{-1}(\sigma)) \epsilon_3 \right. \\ \left. + B_4 \hat{I}_{5/2}(\mathcal{L}^{-1}(\sigma)) (\epsilon_1 + \epsilon_2) \right\} . \quad (30)$$

b. Ferromagnetic Phase

For the FM phase $\cos 2\varphi_i = 1$ and $\sin 2\varphi_i = 0$, thus the single-ion magnetoelastic energy per ion is given by

$$E_{me}^I(\text{FM}) = - \left\{ B_1 \epsilon_3 + B_2 (\epsilon_1 + \epsilon_2) + B_3 \hat{I}_{5/2}(\mathcal{L}^{-1}(\sigma)) \epsilon_3 \right. \\ \left. + B_4 \hat{I}_{5/2}(\mathcal{L}^{-1}(\sigma)) (\epsilon_1 + \epsilon_2) + B_5 \hat{I}_{5/2}(\mathcal{L}^{-1}(\sigma)) (\epsilon_1 - \epsilon_2) \right\} . \quad (31)$$

2. The Two-ion Magnetoelastic Hamiltonian

Two-ion magnetoelastic terms arise from the modulation by the strain of moment-interaction energies such as dipolar, pseudodipolar, and exchange energies. The main contribution to the two-ion magnetoelastic coefficients is from the exchange interaction energy. Conduction-electron redistribution accompanying lattice distortion affects the indirect exchange mechanism, and this change in the indirect exchange energy produces a large two-ion magnetoelastic energy.

The general form for the two-ion contribution to the magnetoelastic Hamiltonian is given by

$$\mathcal{H}_{me}^{II} = -\sum_{f,g} \sum_{i,j,k,\ell=1}^3 D_{ijkl}(f,g) \epsilon_{ij} J_{fk} J_{g\ell} . \quad (32)$$

This term is treated in exactly the same manner as the single-ion term with the obvious analogy $(J_{fi} J_{fj} + J_{fj} J_{fi}) \rightarrow (J_{fi} J_{gj} + J_{fj} J_{gi})$. Thus for the molecular field approximation

$$\begin{aligned} \langle \mathcal{H}_{me}^{II} \rangle = & -\sum_{f,g} \{ D_1(f,g) [\epsilon_1 - \epsilon_2] \langle J_{fx} J_{gx} - J_{fy} J_{gy} \rangle \\ & + D_2(f,g) [\epsilon_1 + \epsilon_2] \langle \vec{J}_f \cdot \vec{J}_g \rangle + D_4(f,g) \epsilon_3 \langle \vec{J}_f \cdot \vec{J}_g \rangle \\ & + 2 D_1(f,g) \epsilon_6 \langle J_{fx} J_{gy} + J_{fy} J_{gx} \rangle . \end{aligned} \quad (33)$$

This expression can be simplified by assuming the dominant contribution to the coefficients $D_n(f,g)$ to be from the exchange interaction energy. With this assumption the strains in the basal plane ϵ_1 , ϵ_2 , and ϵ_6 are expected only to affect significantly I'_0 , the exchange integral for nearest neighbors in the basal plane. Similarly the axial strain ϵ_2 is expected to affect only I'_1 and I'_2 . Therefore we can approximate the two-ion magnetoelastic energy by

$$\begin{aligned} \langle \mathcal{H}_{me}^{II} \rangle = & -\sigma^2 D_5 (\epsilon_1 - \epsilon_2) \sum_i \cos 2\varphi_i \\ & - N \sigma^2 D_4 (\epsilon_1 + \epsilon_2) - 2 \sigma^2 D_5 \epsilon_6 \sum_i \sin 2\varphi_i \\ & - N \sigma^2 (D_1 \cos \varphi + D_2 \cos 2\varphi) \epsilon_3 . \end{aligned} \quad (34)$$

a) Helimagnetic Phase

For the HM phase the $\cos 2\varphi_i$ and $\sin 2\varphi_i$ terms sum to zero, therefore the HM two-ion magnetoelastic energy is given by

$$E_{me}^{II}(HM) = -\sigma^2 D_4 (\epsilon_1 + \epsilon_2) - \sigma^2 (D_1 \cos \varphi + D_2 \cos 2\varphi) \epsilon_3 . \quad (35)$$

b) Ferromagnetic phase

For the FM phase $\cos 2\varphi_i = 1$ and $\sin 2\varphi_i = 0$, therefore the FM two-ion magnetoelastic energy is given by

$$E_{me}^{II}(FM) = -\sigma^2 D_4 (\epsilon_1 + \epsilon_2) - \sigma^2 D_5 (\epsilon_1 - \epsilon_2) - \sigma^2 (D_1 + D_2) \epsilon_3 . \quad (36)$$

3. The Total Magnetoelastic Energy

The two magnetoelastic energy terms may be combined to yield a more compact form by defining the following three functions.

$$f(\sigma) = B_2 + B_4 \hat{I}_{5/2}^{-1}(\sigma) + D_4 \sigma^2 . \quad (37a)$$

$$g(\sigma) = B_1 + B_3 \hat{I}_{5/2}^{-1}(\sigma) . \quad (37b)$$

$$h(\sigma) = B_5 \hat{I}_{5/2}^{-1}(\sigma) + D_5 \sigma^2 . \quad (37c)$$

Using these functions, the total magnetoelastic energy per ion for the HM phase is given by

$$E_{me}^{HM} = -f(\sigma) (\epsilon_1 + \epsilon_2) - g(\sigma) \epsilon_3 - \sigma^2 (D_1 \cos \varphi + D_2 \cos 2\varphi) \epsilon_3 . \quad (38)$$

and the total magnetoelastic energy per ion for the FM phase is given by

$$E_{me}^{FM} = -f(\sigma)(\epsilon_1 + \epsilon_2) - g(\sigma)\epsilon_3 - h(\sigma)(\epsilon_1 - \epsilon_2) - \sigma^2(D_1 + D_2)\epsilon_3 . \quad (39)$$

The Elastic Energy

The fourth term in the Hamiltonian represents the classical elastic energy of the lattice. The general form for $\mathcal{H}_e$ for a crystal of volume V is given by

$$\mathcal{H}_e = \frac{V}{2} \sum_{ijkl} C_{ijkl} \epsilon_{ij} \epsilon_{kl} . \quad (40)$$

Using the symmetry strains, this may be written

$$\mathcal{H}_e = \frac{V}{2} \sum_{\Gamma} \sum_{jj'} C_{jj'}^{\Gamma} \sum_i \epsilon_i^{\Gamma,j} \epsilon_i^{\Gamma,j'} . \quad (41)$$

Thus the symmetry conditions reduce the general expression for the elastic energy per ion to

$$\begin{aligned} E_e = \frac{1}{n} \left[\frac{1}{2} C_{11} \epsilon_1^2 + C_{12} \epsilon_1 \epsilon_2 + C_{13} \epsilon_1 \epsilon_3 \right. \\ \left. + \frac{1}{2} C_{11} \epsilon_2^2 + C_{13} \epsilon_2 \epsilon_3 + \frac{1}{2} C_{33} \epsilon_3^2 \right. \\ \left. + \frac{1}{2} C_{44} \epsilon_4^2 + \frac{1}{2} C_{44} \epsilon_5^2 + (C_{11} - C_{12}) \epsilon_6^2 \right] , \quad (42) \end{aligned}$$

where n is the number density $\frac{N}{V}$, and the C_{ij} 's are the elastic stiffness constants.

The Zeeman Hamiltonian

The last term in the Hamiltonian is the Zeeman Hamiltonian;

$$\mathcal{H}_Z = -g\beta\vec{H} \cdot \sum_i \vec{J}_i . \quad (43)$$

1. Helimagnetic Phase

Neglecting the field-induced distortion of the helix, $\sum_i \langle \vec{J}_i \rangle = 0$, thus the Zeeman energy is zero in the HM phase.

2. Ferromagnetic Phase

In the FM phase for $\vec{H} \parallel \hat{x}$,

$$\langle \mathcal{H}_Z \rangle = -g\beta H \sum_i \langle J_{ix} \rangle . \quad (44)$$

Thus the Zeeman energy per ion in the FM phase is

$$E_Z^{\text{FM}} = -g\beta H J \sigma . \quad (45)$$

II. The Gibbs Free Energy

The generalized Gibbs free energy function is given by⁽⁴⁴⁾

$$G = U + \sum_n F_n X_n - TS, \quad (46)$$

where U is the internal energy, F_n are the generalized forces, X_n are the displacements, and S is the entropy at the temperature T .

For a uniaxial stress P_3 in the $\hat{z}$ direction, the second term in the Gibbs function reduces to $F_z \Delta z$. Consider a disk with an area A and thickness Z , whose volume at $T = 0$ is given by $V = A_0 Z_0$; then

$$F_z \Delta z = P_3 A \Delta z \approx P_3 A_0 \Delta z = V P_3 \epsilon_3. \quad (47)$$

The total entropy consists of two contributions; $S = S_L + S_J$, where S_L is the entropy of the lattice, and S_J is the entropy of the magnetic moment system. Using the molecular field approximation, Bean and Rodbell have shown that S_J is a function only of the reduced magnetization⁽¹¹⁾; that is $S_J = S_J(\sigma)$. The lattice entropy contribution is calculated considering only thermal and external stress distortions of the crystal. Assuming the c -axis linear coef-

ficient of thermal expansion (α_3) and the specific heat at constant volume (C_v) are constant, the lattice entropy for an applied stress along $\hat{z}$ is given by⁽⁴⁵⁾

$$S_L = VE \alpha_3 \epsilon_3 + C_v \ln T, \quad (48)$$

where E is the axial Young's modulus for a hcp crystal

($E \equiv C_{33} - \frac{2C_{13}^2}{C_{11}+C_{12}}$). This is the one-dimensional equivalent of Bean and Rodbell's⁽¹¹⁾ expression $C_v \ln T + \alpha(V-V_0)/K$ for the lattice volume entropy (where α and K are the volume coefficient of thermal expansion and the compressibility respectively). If we define $S(\sigma, T) \equiv [C_v \ln(T) + S_J(\sigma)]/N$, the entropy per ion is given by

$$\frac{S}{N} = \frac{E\alpha_3}{n} \epsilon_3 + S(\sigma, T) \quad (49)$$

where n is the number density $\frac{N}{V}$. Thus the Gibbs free energy per ion is

$$G^{HM(FM)} = E^{HM(FM)} + \frac{P_3}{n} \epsilon_3 - \frac{TE\alpha_3}{n} \epsilon_3 - TS(\sigma, T). \quad (50)$$

The c -axis strain ϵ_3 is defined by

$$\epsilon_3 = \frac{c - c_0}{c_0}, \quad (51a)$$

where c_0 is the value that c would have at $T = 0$ if there were no magnetic or other strains present. c_0 is determined

by plotting c versus temperature above 200 K and extrapolating the straight line obtained in this manner down to 0 K. The slope of this straight line determines α_3 , since $\frac{dc}{dT} = c_0 \frac{d\epsilon_3}{dT} = c_0 \alpha_3$. The strains ϵ_1 and ϵ_2 are defined in a similar manner:

$$\epsilon_1 = \frac{a-a_0}{a_0}, \quad \text{and} \quad \epsilon_2 = \frac{b-b_0}{b_0}. \quad (51b,c)$$

The anomalous strains ϵ'_i due to magnetic effects are obtained by subtracting the thermal contributions from the total strains.

$$\epsilon'_i = \epsilon_i - \alpha_i T. \quad (52)$$

1. HM Phase

Using the expressions for the internal energy per ion and the entropy per ion, the Gibbs free energy per ion in the helimagnetic phase becomes

$$\begin{aligned} G^{HM} = & -[I_0 + (I_1 + D_1 \epsilon_3) \cos \varphi + (I_2 + D_2 \epsilon_3) \cos 2\varphi] \sigma^2 \\ & - f(\sigma) (\epsilon_1 + \epsilon_2) - g(\sigma) \epsilon_3 + E_e + \frac{P_3 \epsilon_3}{n} \\ & - \frac{E \alpha_3 T}{n} \epsilon_3 - TS(\sigma, T). \end{aligned} \quad (53)$$

Minimizing G^{HM} with respect to φ , we obtain

$$\cos \varphi = - \frac{(I_1 + D_1 \epsilon_3)}{4(I_2 + D_2 \epsilon_3)}, \quad \text{or} \quad \epsilon_3 = - \frac{(I_1 + 4I_2 \cos \varphi)}{(D_1 + 4D_2 \cos \varphi)}. \quad (54)$$

Minimizing G^{HM} with respect to ϵ_1 , ϵ_2 , ϵ_4 , ϵ_5 , and ϵ_6 we obtain

$$\epsilon_4^{HM} = \epsilon_5^{HM} = \epsilon_6^{HM} = 0 ,$$

and
$$\epsilon_1^{HM} = \epsilon_2^{HM} = \frac{nf(\sigma)}{C_{11}+C_{12}} - \frac{C_{13}}{C_{11}+C_{12}} \epsilon_3 . \quad (55)$$

Inserting these values into the elastic energy, the elastic energy per ion in the HM phase becomes

$$E_e^{HM} = \frac{E}{2n} \epsilon_3^2 + \frac{nf(\sigma)^2}{C_{11}+C_{12}} . \quad (56)$$

Thus the HM free energy per ion is given by

$$\begin{aligned} G^{HM} = & -[I_0 + I_1 \cos\varphi + I_2 \cos 2\varphi] \sigma^2 + \frac{E}{2n} \epsilon_3^2 \\ & + [-D_1 \sigma^2 \cos\varphi - D_2 \sigma^2 \cos 2\varphi + 2f(\sigma) \frac{C_{13}}{C_{11}+C_{12}} - g(\sigma) \\ & + \frac{P_3}{b} - \frac{E\alpha_3 T}{n}] \epsilon_3 - \frac{nf(\sigma)^2}{C_{11}+C_{12}} - TS(\sigma, T) . \end{aligned} \quad (57)$$

Minimizing G^{HM} with respect to ϵ_3 and using Equation (54), we obtain a cubic equation for $\cos\varphi$.

$$\cos^3\varphi + p \cos^2\varphi + q \cos\varphi + r = 0 ;$$

with $p = \frac{3D_1}{4D_2} ,$

$$q = \left[\frac{D_1^2}{8D_2^2} - \frac{1}{2} + \frac{EI_2}{2n\sigma^2 D_2^2} - \frac{f(\sigma)C_{13}}{\sigma^2 D_2 (C_{11}+C_{12})} - \frac{g(\sigma)}{2D_2\sigma^2} \right]$$

$$- \frac{P_3}{2n\sigma^2 D_2^2} + \frac{E\alpha_3 T}{2n\sigma^2 D_2^2} \Big] ,$$

$$\text{and } r = \frac{D_1}{8D_2^2} \left[\frac{EI_1}{D_1 n\sigma^2} - \frac{2f(\sigma)C_{13}}{(C_{11}+C_{12})\sigma^2} + \frac{g(\sigma)}{\sigma^2} - D_2 + \frac{E\alpha_3 T - P_3}{n\sigma^2} \right] . \quad (58)$$

The expression for G^{HM} becomes quite complicated when expressed as a function of the phenomenological constants, σ , and the macroscopic parameters (P_3 , T , α_3 , n , E , and the C_{ij} 's); therefore, we will use the expression

$$G^{\text{HM}} = -[I_0 + I_1 \cos\varphi + 2I_2 \cos^2\varphi - I_2] \sigma^2 - \frac{nf(\sigma)^2}{(C_{11} + C_{12})} - \frac{E}{2n} \epsilon_3^{\text{HM}^2} - TS(\sigma, T) , \quad (59)$$

where the equilibrium c-axis strain ϵ_3^{HM} is given by

$$\epsilon_3^{\text{HM}} = \frac{nD_1\sigma^2}{E} \cos\varphi + \frac{2nD_2\sigma^2}{E} \cos^2\varphi - \frac{nD_2\sigma^2}{E} - \frac{2nf(\sigma)C_{13}}{E(C_{11} + C_{12})} + \frac{ng(\sigma)}{E} - \frac{P_3}{E} + \alpha_3 T , \quad (60)$$

and $\cos\varphi$ is given by Equation (58).

2. FM Phase

The Gibbs free energy per ion in the ferromagnetic phase is given by

$$\begin{aligned}
G^{\text{FM}} = & -[I_0 + I_1 + D_1 \epsilon_3 + I_2 + D_2 \epsilon_3] \sigma^2 - h(\sigma) (\epsilon_1 - \epsilon_2) \\
& - f(\sigma) (\epsilon_1 + \epsilon_2) - g(\sigma) \epsilon_3 + E_e + \frac{P_3 \epsilon_3}{n} \\
& - \frac{E \alpha_3 T}{n} \epsilon_3 - g \beta H J \sigma - TS(\sigma, T) .
\end{aligned} \tag{61}$$

Minimizing G^{FM} with respect to ϵ_1 , ϵ_2 , ϵ_4 , ϵ_5 , and ϵ_6 we obtain

$$\epsilon_4^{\text{FM}} = \epsilon_5^{\text{FM}} = \epsilon_6^{\text{FM}} = 0 , \tag{62a}$$

$$\epsilon_1^{\text{FM}} - \epsilon_2^{\text{FM}} = \frac{2nh(\sigma)}{C_{11} - C_{12}} , \tag{b}$$

$$\epsilon_1^{\text{FM}} = \frac{nh(\sigma)}{C_{11} - C_{12}} + \frac{nf(\sigma)}{C_{11} + C_{12}} - \frac{C_{13}}{C_{11} + C_{12}} \epsilon_3 , \tag{c}$$

$$\text{and } \epsilon_2^{\text{FM}} = -\frac{nh(\sigma)}{C_{11} - C_{12}} + \frac{nf(\sigma)}{C_{11} + C_{12}} - \frac{C_{13}}{C_{11} + C_{12}} \epsilon_3 . \tag{d}$$

Inserting these values into the elastic energy, the elastic energy per ion in the FM phase becomes

$$E_e^{\text{FM}} = \frac{E}{2n} \epsilon_3^2 + \frac{nh(\sigma)^2}{C_{11} - C_{12}} + \frac{nf(\sigma)^2}{C_{11} + C_{12}} . \tag{63}$$

Thus the FM free energy per ion is given by

$$\begin{aligned}
G^{\text{FM}} = & -[I_0 + I_1 + I_2] \sigma^2 + \frac{E}{2n} \epsilon_3^2 \\
& + [-D_1 \sigma^2 - D_2 \sigma^2 + \frac{2f(\sigma)C_{13}}{C_{11} + C_{12}} - g(\sigma) + \frac{P_3}{n} - \frac{E \alpha_3 T}{n}] \epsilon_3 \\
& - \frac{nh(\sigma)^2}{C_{11} - C_{12}} - \frac{ng(\sigma)^2}{C_{11} + C_{12}} - g \beta H J \sigma - TS(T, \sigma) .
\end{aligned} \tag{64}$$

Minimizing G^{FM} with respect to ϵ_3 , we obtain

$$G^{\text{FM}} = -[I_0 + I_1 + I_2] \sigma^2 - \frac{nh(\sigma)^2}{(C_{11} - C_{12})} - \frac{nf(\sigma)^2}{C_{11} + C_{12}} \\ - \frac{E}{2n} \epsilon_3^{\text{FM}^2} - g\beta H J \sigma - TS(T, \sigma) , \quad (65)$$

where the equilibrium c-axis strain ϵ_3^{FM} is given by

$$\epsilon_3^{\text{FM}} = \frac{n}{E} (D_1 + D_2) \sigma^2 - \frac{2nf(\sigma)C_{13}}{E(C_{11} + C_{12})} + \frac{nq(\sigma)}{E} \\ - \frac{P_3}{E} + \alpha_3 T . \quad (66)$$

III. The Critical Field

The transition from the helimagnetic structure to the ferromagnetic structure occurs at the critical field H_c which causes the FM Gibbs free energy per ion to equal the HM Gibbs free energy per ion. Thus

$$g\beta J\sigma H_c = -[I_1(1-\cos\varphi) + 2I_2(1-\cos^2\varphi)]\sigma^2 - \frac{nh(\sigma)^2}{(C_{11}-C_{12})} + \frac{E}{2n} (\epsilon_3^{HM^2} - \epsilon_3^{FM^2}), \quad (67)$$

or

$$H_c = \frac{\sigma}{g\beta J} \left\{ -[I_1(1-\cos\varphi) + 2I_2(1-\cos^2\varphi)] - \frac{nh(\sigma)^2}{\sigma^2(C_{11}-C_{12})} - \frac{n}{2E} [D_1\sigma^2(1+\cos\varphi) + 2D_2\sigma^2\cos^2\varphi] - \frac{4f(\sigma)C_{13}}{C_{11}+C_{12}} + 2g(\sigma) - \frac{2P_3}{n} + \frac{2E\alpha_3 T}{n}] [D_1(1-\cos\varphi) + 2D_2(1-\cos^2\varphi)] \right\}. \quad (68)$$

The change in the critical field caused by an applied stress along the "c" axis is given by

$$\begin{aligned} \Delta H_c &= H_c(P_3) - H_c(0) \\ &= \frac{\sigma}{g\beta J E} \left\{ [I_1 E - nD_1 D_2 \sigma^2 + 2nD_1 f(\sigma) \frac{C_{13}}{(C_{11}+C_{12})} - nD_1 g(\sigma) - D_1 E \alpha_3 T] (\cos\varphi - \cos\varphi_0) \right\} \end{aligned}$$

$$\begin{aligned}
& + [2I_2E + \frac{n}{2} D_1^2 \sigma^2 - 2nD_2^2 \sigma^2 + 4nD_2 f(\sigma) \frac{C_{13}}{C_{11}+C_{12}} \\
& - 2nD_2 g(\sigma) - 2D_2 E \alpha_3 T] (\cos^2 \varphi - \cos^2 \varphi_0) \\
& + 2nD_1 D_2 \sigma^2 (\cos^3 \varphi - \cos^3 \varphi_0) \\
& + 2nD_2^2 \sigma^2 (\cos^4 \varphi - \cos^4 \varphi_0) \\
& + P_3 [D_1 (1 - \cos \varphi) + 2D_2 (1 - \cos^2 \varphi)] \} \tag{69}
\end{aligned}$$

where $\varphi = \varphi(P_3)$ and $\varphi_0 = \varphi(0)$. Experimental results and the cubic equation for $\cos \varphi$ suggest that $\varphi \approx \varphi_0$ for small stresses. Thus the shift in the critical field caused by P_3 can be approximated by

$$\Delta H_c = \frac{\sigma P_3}{g\beta J E} [D_1 (1 - \cos \varphi_0) + 2D_2 (1 - \cos^2 \varphi_0)] . \tag{70}$$

This indicates that ΔH_c is linearly dependent on the stress P_3 and that the two-ion magnetoelastic interaction is the dominant source of the change in the internal energy caused by uniaxial stress along "c." The temperature dependence of the slope $\Delta H_c / \Delta P_3$ is determined by the temperature dependence of σ and φ_0 ; thus, experimental measurements of $\Delta H_c / \Delta P_3$, σ , φ_0 , and E should yield an accurate determination of the two-ion magnetoelastic coefficients D_1 and D_2 .

APPARATUS AND EXPERIMENTAL PROCEDURE

The objective was to measure the shift in the critical field as a function of an applied uniaxial stress in the "c" direction. For a slowly increasing d.c. magnetic field with a sinusoidal modulation, the current developed in a coil with the sample acting as its core is proportional to

$$\frac{dH}{dt} = \frac{dH_{dc}}{dt} + \omega H_{ac} \cos \omega t + \frac{dM(T,H)}{dt} . \quad (71)$$

The magnetic susceptibility is given by $\chi(T,H) = \frac{dM(T,H)}{dH}$,

thus

$$\frac{dH}{dt} = \frac{dH_{dc}}{dt} + \omega H_{ac} \cos \omega t + \alpha \chi(T,H) \frac{dH_{dc}}{dt} + \alpha \chi(T,H) \omega H_{ac} \cos \omega t . \quad (72)$$

A dummy coil in the same magnetic environment as the sample coil develops a current proportional to

$$\frac{dH'}{dt} = \frac{dH_{dc}}{dt} + \omega H_{ac} \cos \omega t . \quad (73)$$

By subtracting the dummy coil signal from the sample coil signal, we obtained an a.c. current (I) with an amplitude proportional to the susceptibility.

$$I = \chi(T,H) \gamma \omega \cos \omega t \quad (74)$$

for $\frac{dH_{dc}}{dt} \ll \omega H_{ac}$. This signal was then amplified, synchronously rectified, and filtered to provide a signal proportional to the susceptibility which drove the y-axis of an x-y recorder. The x-axis was driven by the output of a direct reading Hall-effect gaussmeter, so that a plot of susceptibility versus field was obtained. The abrupt increase in magnetization at the critical field gives rise to a maximum in the susceptibility. Uniaxial stress of varying magnitude was applied to the sample, and the corresponding shift of the susceptibility maximum as a function of applied stress could then be easily measured.

I. Sample Preparation

In order to apply a magnetic field in the basal plane and a uniaxial stress along the "c" axis, c-plane platelets were spark-cut from a dysprosium single crystal obtained from Metals Research Ltd., Cambridge, England. The quoted purity of the crystal was 99.9%; however, no tests were conducted on the sample to verify this. The crystal was in the form of a cylinder 6mm in diameter and 25mm long grown by the inductive-heating floating-zone technique. Laue back-reflection x-ray examination indicated that the angle between the symmetry axis of the cylinder and the "c" axis

of the crystal was 19° ; thus, the maximum basal-plane dimension of a c-plane platelet was limited to approximately 6mm. The demagnetizing factor for the center of a short cylinder, magnetized perpendicular to its symmetry axis, is given by

$$D = 2\pi \frac{T}{\sqrt{d^2 + T^2}}, \quad (75)$$

where T is the thickness and d the diameter of the cylinder. Thus the internal field is given by

$$H_{\text{int}} = H_{\text{applied}} - 2\pi \frac{T}{\sqrt{d^2 + T^2}} M(T, H). \quad (76)$$

In order to measure the critical field accurately, it is important to have as small a demagnetizing factor as possible, since the magnetization $M(T, H)$ is changing most rapidly near the critical field. It is therefore necessary that T/d be made as small as possible. The signal strength for the susceptibility measurement decreases, however, with decreasing sample volume; thus, for a fixed maximum diameter, the minimum thickness is dictated by the signal-to-noise ratio. Furthermore, for thicknesses less than .1mm the crystals become extremely fragile. Consequently, a c-plane platelet .25mm thick was spark-cut from the stock crystal, spark-planed down to a thickness of .18mm, and machined to a cylindrical shape. This size gave a satisfactory signal-to-

noise ratio with suitable amplification and filtering. A larger signal-to-noise ratio would be desirable, but this size corresponds to a demagnetizing factor $D = .17586$ which typically causes a shift of 200 Oe in the observed critical field (assuming $M(T, H_c) = \frac{1}{2} M_s(T)$, where $M_s(T)$ is the saturation value of magnetization at the temperature T).

II. Cryogenic System

In order to perform susceptibility measurements in the HM temperature range, a suitable means of maintaining the sample at any desired temperature between 85 K and 179 K with good temperature stability is needed. For this purpose, a cryogenic dewar system (Andonian 0/25/200/7M) was used. The main dewar body contains a 3 liter liquid helium reservoir with a 4 liter liquid nitrogen heat shield. The long cylindrical sample chamber extends through the main dewar body into a narrow tail section. The lower portion of the sample chamber, together with its vacuum jacket, could be lowered into the center of the gap of our Harvey Wells Model L-128 magnet.

The sample and stress mechanism was mounted on a thin-walled stainless steel support tube and inserted from the top of the system. Helium is throttled to the sample zone through a heat exchanger and diffuser and flows around and

in intimate contact with the sample. Temperature gradients are extremely small so that accurate temperature measurements can be made. The throttle valve controls the flow of helium and consequently the rate of energy removal from the sample zone. The cooling capacity of the exhaust gas is used to remove thermal energy conducted down the sample support and wire leads. Since the sample zone is isolated from the refrigerant reservoir, high sample temperatures are achieved with little increase in refrigerant consumption. Exhaust-gas flow along the top-loading sample holder permits rapid sample change without contamination of system or sample. Only a few minutes need elapse between measurements on successive samples. Liquid-helium consumption depended upon the number of different equilibrium temperatures used on a run but averaged approximately .1 liter per hour.

To monitor and control the temperature, a copper-constantan thermocouple was used. One junction was mounted on the stress mechanism near the sample. Another junction was placed in a liquid nitrogen reference bath. The emf's thus produced were measured with a Leeds and Northrup K-3 potentiometer in conjunction with a Fluke Model 840A electronic galvanometer. The output from the galvanometer was amplified by a Philbrick USA-3 operational amplifier with a

high input impedance to prevent large current drains from the galvanometer. The output voltage from the operational amplifier was used to drive a Kepco Model CK36-1.5M power supply in a remote programming mode. A diode placed across the amplifier output limits the negative voltage applied to the bridge in the power supply control circuit to about .6 volts, preventing damage to that circuit. The power supply provides power to a heater on the sample mount, so that if a small temperature decrease is detected, the feedback system drives the heater with a proportional correction. A slight excess rate of cooling is established by adjustment of the helium throttle valve; then the feedback system supplies the appropriate electrical power to attain equilibrium at the desired temperature.

The heater consisted of a constantan wire wound bifilarly around the brass stress mechanism directly beneath the sample. The total resistance of the heater was fifty ohms, providing a maximum power input to the sample mount of 4.5 watts. A variable feedback resistor on the operational amplifier provided a means of obtaining the best temperature stabilization. At equilibrium, the heater operated at a few milliwatts average power, depending on the equilibrium temperature. The temperature remained constant to within

TEMPERATURE CONTROL SYSTEM
FIGURE 8

± 15 millidegrees at equilibrium.

III. Stress Mechanism and Sample Support

A simple screw, lever, and piston mechanism was used to apply uniaxial stress to the sample. The main portion was machined from a 25.4mm diameter nonmagnetic brass cylinder 50mm long. A 6.3mm diameter cylindrical hole 25mm long constrained the piston to move perpendicular to the sample support surface. The piston consisted of a 6.3mm diameter polished single-crystal ruby rod 25mm long cemented to a 6.3mm diameter brass rod 6mm long with a conical tip. The brass cone distributed the force exerted by the lever, and the 25mm ruby crystal insured a uniform stress on the sample. (No tests were conducted, however, to verify that the stress was uniform.) The lever was a 6.3mm x 6.3mm x 16mm rectangular brass arm pivoted on a 3.15mm diameter stainless steel pin to give a 3:1 mechanical advantage. A 4.8mm diameter stainless steel rod extending down through the top of the sample support was used as a screw to apply force to the lever.

It is important for this type of measurement that the "a" and "b" axes not be constrained. Any applied stresses in the basal plane of the sample will affect the equilibrium

STRESS MECHANISM AND SAMPLE SUPPORT

FIGURE 10

c-axis strain, leading to erroneous results. With this in mind, the samples were mounted in the following manner. Two pieces of loose weave silk cloth were soaked in Duco cement and placed on each surface of a 2mm thick fused quartz pad, and the sample was placed on top of one silk covered surface. The sample and pad were then placed on the brass support, and a moderate stress was applied to the sample with the ruby piston during the drying period. The surface of the ruby piston was polished flat; however, a thin sheet of teflon was placed between the ruby and the sample to compensate for surface irregularities in the sample. This technique was expected to insure that the sample surface was exactly parallel to the surface of the ruby piston since any small misalignment would presumably be corrected in the silk and cement layers. The thick silk and cement layer attaching the sample to the quartz pad is flexible enough, even at low temperatures, to ensure stress free magnetostriction in the basal plane.

The stress was calculated from the observed strain in the ruby piston. A paper-backed constantan foil SR-4 strain gauge (FAP-06-12), manufactured by Baldwin-Lima-Hamilton, was bonded to the side of the ruby crystal parallel to its symmetry axis with Duco cement. Thermal and magnetoresistive compensation was achieved by cementing a dummy gauge to

a second ruby crystal mounted on top of the stress mechanism, thus it was in the same thermal and magnetic environment as the active gauge. These two gauges were electrically connected in adjacent arms of a Wheatstone bridge incorporated in a Baldwin-Lima-Hamilton Model 120 direct reading strain indicator. The advantages of the constantan foil over other gauge materials stem from its small temperature dependence of resistance and gauge factor. A disadvantage of the constantan foil is its relatively large magnetoresistance at low temperatures. Probably because of misalignment of the dummy gauge, magnetoresistive effects were not negligible; consequently, all strain measurements were taken with zero applied field.

The stress ($P_3 = \tau_3'$) was calculated from the observed strain (ϵ_3') by the stress-strain relation $\epsilon_i' = S_{ij}' \tau_j'$, where the S_{ij}' 's are the elastic compliances of ruby in the coordinate system of the rod ($\hat{z}' \parallel$ symmetry axis). For the specific case of uniaxial stress in the $\hat{z}'$ direction, this reduces to $\epsilon_{33}' = S_{33}' P_3$. In the crystal coordinate system ($\hat{z} \parallel c$, $\hat{x} \parallel a$, and $\hat{y} \parallel b$), using symmetry considerations, the elastic compliance tensor for ruby takes the form

$$s_{ij} = \begin{bmatrix} s_{11} & s_{12} & s_{13} & s_{14} & 0 & 0 \\ s_{12} & s_{11} & s_{13} & -s_{14} & 0 & 0 \\ s_{13} & s_{13} & s_{33} & 0 & 0 & 0 \\ s_{14} & -s_{14} & 0 & s_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & s_{44} & 2s_{14} \\ 0 & 0 & 0 & 0 & 2s_{14} & 2(s_{11}-s_{12}) \end{bmatrix} \quad (77)$$

Laue back-reflection x-ray examination indicated the relationship between the two coordinate systems as in Figure 11.

Figure 11

where $\theta = 19^\circ$ and $\phi = 15^\circ$. This is an Euler transformation with $\psi = 0$. This transformation yields a relation between s'_{33} and the elastic compliances measured in the crystallographic coordinate system.

$$S'_{33} = \sin^4 \theta S_{11} + \cos^4 \theta S_{33} + \sin^2 \theta \cos^2 \theta S_{44} \\ + 2 \sin^2 \theta \cos^2 \theta S_{13} - 2 \sin^3 \theta \cos \theta \cos \varphi (3 \sin^2 \varphi - \cos^2 \varphi) S_{14} .$$

The following values for Al_2O_3 were used⁽⁴⁶⁾:

$$\begin{aligned} S_{11} &= 2.353 \pm 0.002 \times 10^{-13} \text{ cm}^2/\text{dyne}, \\ S_{33} &= 2.170 \pm 0.002 \times 10^{-13} \text{ cm}^2/\text{dyne}, \\ S_{44} &= 6.940 \pm 0.008 \times 10^{-13} \text{ cm}^2/\text{dyne}, \\ S_{12} &= -.716 \pm 0.007 \times 10^{-13} \text{ cm}^2/\text{dyne}, \\ S_{13} &= -.364 \pm 0.006 \times 10^{-13} \text{ cm}^2/\text{dyne}, \text{ and} \\ S_{14} &= 0.489 \pm 0.005 \times 10^{-13} \text{ cm}^2/\text{dyne}. \end{aligned} \quad (78)$$

This yields $S'_{33} = 2.372 \times 10^{-13} \text{ cm}^2/\text{dyne}$, or

$$P_3 = 4.216 \times 10^{12} \epsilon'_3 \text{ dyne/cm}^2. \quad (79)$$

IV. Detector and Dummy Coils

Two coils of A.W.G. 48 ($d = .0318\text{mm}$) copper magnet wire were wound on $1.6\text{mm} \times 6.35\text{mm} \times 9.5\text{mm}$ plexiglass cores with $.8\text{mm} \times 12.7\text{mm} \times 12.7\text{mm}$ nylon sides. They each had approximately 5000 turns, giving an inductance of 500 millihenries each. These were connected in series and cemented in the recesses provided for them on the stress mechanism with their centers aligned such that the sample was positioned half-way between the coils on a line connecting their centers.

These coils have the maximum number of turns, hence the largest current output, that could be used in the limited space of the sample chamber.

A dummy coil, consisting of 3000 turns of A.W.G. 40 ($d = .08\text{mm}$) copper magnet wire, was wound on a cylindrical nylon coil bobbin (I.D. = 9.5mm, O.D. = 19mm, and 9.5mm wide).

V. Electronic Equipment

The magnetic field was produced by a Harvey Wells Model L-128 Magnet with a 5cm pole-piece gap, equipped with a set of modulation coils. The magnet power supply (Harvey Wells Model HS-1050) was equipped with a mechanical sweep capable of increasing (or decreasing) the d.c. field at various uniform sweep rates to a maximum field of 18 kOe. The modulation coils were driven by a Dynaco Mark III 60 watt audio amplifier. Optimum results were achieved for a d.c. field increase rate of 10 Oe/second and a sinusoidal modulation field 90 Oe peak-to-peak at 15 Hz. The signal-to-noise ratio was a maximum for the largest modulation attainable. With the Dynaco amplifier operating at its maximum undistorted output, the maximum modulation was achieved at 15 Hz for the modulation coils used.

The magnetic field was measured with a Bell Model 240 gaussmeter with a Model HFT2401 high field Hall probe, accurate to 2%. The gaussmeter output was filtered to remove the a.c. portion of the field, and the resulting d.c. signal drove the x axis of a Moseley 7001AM x-y recorder.

The dummy coil was mounted on the face of one pole piece of the magnet, and its output was connected to a solid-state variable phase shifter, capable of continuously varying the phase of the signal from 0° to slightly greater than 180° . A Union Carbide Model H6010 solid-state operational amplifier was used to sum the signals from the detector and dummy coils. The phase and amplitude of the dummy coil were adjusted at each temperature to minimize the signal output of the operational amplifier for an applied field less than $H_c(T)$. A Princeton-Applied-Research Model JB-4 lock-in amplifier in its internal reference mode supplied the input signal to the Dyna power supply and provided signal gain to drive the y axis of the x-y recorder. This amplifier utilizes narrow bandwidth phase sensitive detection to convert a low level a.c. signal into a noise-free d.c. output.

BLOCK DIAGRAM OF EXPERIMENTAL APPARATUS

FIGURE 12

RESULTS AND DISCUSSION

The critical field transitions characteristically produce a susceptibility maximum and, therefore, a peak in the current produced by the coil containing the sample. This peak was found to be sharp for no applied stress and reversible with d. c. magnetic field sweep direction for slow sweep rates. The line width at half maximum was typically 700 Oe for zero stress measurements, becoming smaller with decreasing temperature. Assuming the magnetization at H_c to be one-half the saturation magnetization, the zero-stress critical field maxima were corrected for the demagnetizing field and plotted as a function of internal field. The critical field versus temperature curve thus obtained yields reasonable agreement with critical field measurements determined by other methods.

Uniaxial stress applied along the "c" axis caused the susceptibility maxima to be shifted to higher fields. The susceptibility curves shown in Figure 14 at 155 K illustrate the general characteristics observed at all temperatures. The application of stress causes a broadening of the susceptibility peak and a decrease in the maximum value of the susceptibility. The broadening effect is temperature independent and increases with increasing stress causing a line

FIGURE 13

FIGURE 14

width at half maximum of approximately 1500 Oe for a stress of 500 bar. The maximum value of the susceptibility decreases with increasing stress; and this effect is temperature dependent, being most pronounced at low temperature. The combined effect of peak broadening and suppression causes difficulty in determining the critical field for large stresses; therefore, the measurements were limited to a maximum stress of 600 bar. A depression of the susceptibility peak was also observed by Milton and Scott⁽¹⁷⁾ in their measurements of the hydrostatic pressure dependence of the Curie temperature in polycrystalline dysprosium.

Magnetization measurements⁽¹⁾ indicate that the magnitude of the susceptibility at the critical field should increase with decreasing temperature. The pick-up coil current peak increased in magnitude with decreasing temperature down to approximately 130 K and then decreased for lower temperatures. This anomalous behavior is probably associated with the temperature dependence of the pick-up coil characteristics. Due to this effect and the peak suppression caused by the applied stress, critical field shift measurements became impossible for temperatures below 110 K. Since the theoretical approximation ignoring the basal plane anisotropy is not valid below 110 K, no effort was made to correct this

deficiency of the experimental apparatus.

The shift in the critical field as a function of applied c-axis stress was measured in the temperature range 120-170 K. At all temperatures in this range, the critical field increased linearly with increasing stress. The critical field shift ΔH_c was plotted as a function of the stress ΔP_3 , and the slope $\frac{\Delta H_c}{\Delta P_3}$ was determined by using the least squares technique.

Equation (70) may be written

$$\frac{\Delta H_c}{\Delta P_3} = \frac{\sigma}{g\beta JE} [D_1(1-\cos\varphi_0) + 2D_2(1-\cos^2\varphi_0)] \quad (70)$$

The axial Young's modulus E can be calculated from the elastic constants C_{33} , C_{13} , C_{12} , and C_{11} ($E = C_{33} - \frac{2C_{13}^2}{C_{11}+C_{12}}$). Figure 16 is a plot of the elastic constants of dysprosium taken from Fisher and Dever's⁽⁴⁷⁾ data for high temperatures and extrapolated down to low temperatures. E varies from $.7000 \times 10^{12}$ dyne/cm² at 110 K to $.7004 \times 10^{12}$ dyne/cm² at T = 170 K, therefore it can be considered a constant; $E = .7 \times 10^{12}$ dyne/cm². The experimental values of $\varphi_0(T)$ and $\sigma(T)$ are taken from Figures 2 and 3. For $g = 4/3$ and $J = 15/2$, a best fit to $\frac{\Delta H_c}{\Delta P_3}$ versus T data was obtained when

$$\begin{aligned}
D_1 &= 2.00 \times 10^{-11} \text{ ergs} = 140 \times 10^3 \text{ K} \\
D_2 &= -.514 \times 10^{-11} \text{ ergs} = -37 \times 10^3 \text{ K}
\end{aligned} \tag{80}$$

The theoretical temperature dependence of $\frac{\Delta H_c}{\Delta P_3}$ using these values for D_1 and D_2 is compared with the experimental data in Figure 17.

Equation (70) can be derived from a simple Gibb's free energy per ion introduced by Landry⁽⁴⁵⁾ to explain the temperature variation of the helical turn angle.

$$\begin{aligned}
G^L &= -[I_0 + (I_1 + D_1 \epsilon_3) \cos \varphi + (I_2 + D_2 \epsilon_3) \cos 2\varphi] \sigma^2 \\
&\quad + \frac{E}{2n} \epsilon_3^2 - \frac{TE\alpha_3 \epsilon_3}{n} + \frac{P_3 \epsilon_3}{n} .
\end{aligned} \tag{81}$$

Equation (81) is equivalent to Landry's Equation (2) for the Gibb's free energy per unit volume, where $J_i^L = nI_i$, $D_i^L = nD_i$, $w^L = \varphi$, $\lambda_c^L = \epsilon_3$, and the subscript $c = 3$. Landry obtained a best fit to c versus T data when

$$\begin{aligned}
\frac{D_1^L}{n} &= .996 \times 10^{-13} \text{ ergs} = 720 \text{ K} , \\
\frac{D_2^L}{n} &= .462 \times 10^{-13} \text{ ergs} = 335 \text{ K} .
\end{aligned} \tag{82}$$

FIGURE 15

ELASTIC CONSTANTS OF DYSPROSIUM
FIGURE 16

FIGURE 17

The c versus T curve is generated from the equilibrium helimagnetic c -axis strain ϵ_3^{HM} . Using Landry's simple Hamiltonian ϵ_3^{HM} is given by

$$\epsilon_3^{\text{L}} = \frac{\sigma^2}{E} [nD_1 \cos\varphi + nD_2 \cos 2\varphi] - \frac{P_3}{E} + \alpha_3 T \quad (83)$$

This is to be compared with Equation (60).

$$\begin{aligned} \epsilon_3^{\text{HM}} = \frac{\sigma^2}{E} [nD_1 \cos\varphi + nD_2 \cos 2\varphi] - \frac{P_3}{E} + \alpha_3 T \\ - \frac{2nf(\sigma)C_{13}}{E(C_{11} + C_{12})} + \frac{ng(\sigma)}{E} . \end{aligned} \quad (60)$$

where $f(\sigma)$ and $g(\sigma)$ are given by

$$\begin{aligned} f(\sigma) &= B_2 + B_4 \hat{I}_{5/2}(\mathcal{L}^{-1}(\sigma)) + D_5 \sigma^2 \\ g(\sigma) &= B_1 + B_3 \hat{I}_{5/2}(\mathcal{L}^{-1}(\sigma)) \end{aligned} \quad (37)$$

Thus the large discrepancy in the values for D_1 and D_2 could be due to the fact that Landry ignored the single-ion magnetoelastic energy when fitting the c versus T data.

The values for D_1 and D_2 given in Equation (80) are not accurately determined by the simplified theory presented in this thesis. A definite relationship between D_1 and D_2 is, however, accurately determined. Figure 18 is a plot of D_1 versus D_2 for several temperatures using Equation (70).

FIGURE 18

D_1 and D_2 would be uniquely determined if the straight lines, representing the relationship of D_1 to D_2 , all had a common intersection point. Figure 18 does indicate that, for any given value of D_2 , D_1 is determined to within a few per cent. A more accurate experimental determination of $\Delta H_c / \Delta P_3$ as a function of temperature and the inclusion of the Zeeman energy in the theory for the helimagnetic phase would hopefully remove the uncertainty in the unique values of D_1 and D_2 .

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ACKNOWLEDGEMENTS

I would like to express my appreciation to Dr. P. L. Donoho, under whose supervision this work was carried out, for his guidance and his many helpful suggestions. Also, I would express my appreciation to my colleagues in the rare-earth laboratory for their assistance.

A special note of thanks is due Mrs. Mary Comerford for typing this thesis. Rice University, the National Science Foundation, and the National Aeronautics and Space Administration are acknowledged for their financial support. I especially wish to thank the Department of Health, Education and Welfare for an N.D.E.A. traineeship. The helium used in this experiment was supplied by the Office of Naval Research.

Last, but not least, I would express my appreciation to my wife, Donna, for her patience and constant encouragement without which this work would not have been possible.