Magnetoelastic properties of dysprosium aluminum garnet: Theory

W. P. Wolf
Becton Center, Yale University, New Haven, Connecticut 06520

C. H. A. Huan
Clarendon Laboratory, Oxford University, Oxford OX1 3PU, United Kingdom
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Expressions are derived for the first- and second-order magnetoelastic-energy terms for the Ising-like antiferromagnet dysprosium aluminum garnet. The first-order terms are used to interpret the field dependence of the magnetostriction, and the second-order terms for a similar analysis of recent measurements of magnetoacoustic effects. The agreement is generally satisfactory. The theory also predicts some unusual piezomagnetic effects, one of which may explain the anomalous neutron scattering near the phase boundary in fields parallel to [001] and [110]. The results of this work suggest that magnetoelastic effects may be important for the detailed understanding of large classes of magnetic materials, characterized by an antiferromagnetic structure which does not enlarge the unit cell.

I. INTRODUCTION

Dysprosium aluminum garnet (DyAlG) is an Ising-like antiferromagnet, whose magnetic, thermal, and optical properties have been studied extensively.[1-4] In most of these studies, the magnetoelastic effects were ignored, largely because DyAlG is extremely stiff [Debye $\Theta_D = 500$ K (Ref. 1)] and the magnetic interactions are relatively weak, corresponding to a Néel temperature $T_N \approx 2.5$ K. Only in connection with some mangeto-optical experiments, in which a surprising birefringence was observed,[5] were the effects of magnetoelastic distortions invoked to explain the initially unexpected effects. Subsequent measurements of magnetostriction[6,7] confirmed the importance of magnetoelastic distortions and a study of the effect of applying magnetic fields in different directions was found to be generally in agreement with qualitative predictions based on symmetry considerations.[7]

In this paper we develop a more detailed theory, also based on symmetry considerations, which will be useful for explaining both the low- and high-field magnetoelastic effects which are observed. The theory will be asymptotically exact at high fields and at low temperatures, but it should provide a good qualitative model for all fields and temperatures.

By extending the theory to include terms to second order in the various strain components, we can make similar predictions of various magnetoacoustic effects. Recent experiments on DyAlG showed a number of striking variations of sound velocity with magnetic field,[8] which had no ready explanation, although singularities in the curves clearly corresponded to magnetic phase transitions, well known from previous studies.[9] Our theory will explain the general features observed and provide an overall structure for analyzing more extensive magnetoacoustic studies which are now in progress.[10]

The theory also provides some insight into more conventional magnetic properties which may be affected by magnetoelastic effects. For example, the theory shows that a shear strain will lead to a magnetic moment in zero field when the material is in the antiferromagnetic state. This unusual form of piezomagnetism may explain some of the anomalous low-field behavior which has been observed in polycrystalline samples.[11,12]

It seems clear that magnetoelastic effects can be important for the detailed understanding not only of DyAlG but also of other materials, especially those in which the magnetic order parameter corresponds to zero wave vector. In all such systems, couplings between the strain, the magnetic order, and the bulk magnetization may be allowed by symmetry, and corresponding magnetoelastic effects may occur. We shall not address these more general considerations in any detail, but it should become clear that the effects we discuss for the case of DyAlG are by no means restricted to this relatively complex case.

In Sec. II we shall present the general theory, which involves listing the terms in the magnetoelastic energy allowed by symmetry and deducing the corresponding magnetostrictive and magnetoacoustic changes produced by fields applied in various directions. In Sec. III we shall then apply some of these results, first to explain some of the magnetostrictive effects observed by Dillon *et al.*[6,7,13] and then to the recently observed magnetoacoustic effects.[8] In Sec. III we shall also make estimates of piezomagnetic effects which may occur in this material. Various microscopic mechanisms for all of the magnetoelastic terms are discussed in Sec. IV. In Sec. V we give a brief discussion of the relevance of these effects for other experiments and other materials.

II. THEORY

A. Magnetic order parameters

Dysprosium aluminum garnet has a relatively complex structure with 12 spins per unit cell.[14,15] Corresponding-

ly, there are 36 different order parameters which may be involved in the description of an arbitrary state of magnetic order indexed within the crystallographic unit cell. These order parameters have been classified according to their symmetry properties by Mukamel and Blume[16] (MB), whose notation we shall generally follow here.

Fortunately, the Ising-like behavior of the Dy^{3+} spins in DyAlG reduces significantly the number of order parameters needed to describe any magnetic state of this system to a high degree of approximation. There are six. Following the notation of MB, we write

$$M_{2\mu}=\sum_{i\alpha} S^{\mu}_{\mu i\alpha}N_{\mu} \quad (\mu=x,y,z) , \tag{1a}$$

$$\eta=\sum_{\mu,i\alpha} \alpha S^{\mu}_{\mu i\alpha}N_{\mu}/3 \quad (\mu=x,y,z) , \tag{1b}$$

$$\gamma_{x^2}=-\sum_{i\alpha} \alpha S^{y}_{yi\alpha}N_{y}/2+\sum_{1\alpha} \alpha S^{z}_{zi\alpha}N_{z}/2 , \tag{1c}$$

$$\gamma_{y^2}=\sum_{i\alpha} \alpha S^{x}_{xi\alpha}N_{x}/2-\sum_{1\alpha} \alpha S^{z}_{zi\alpha}N_{z}/2 , \tag{1d}$$

where we have introduced normalizing factors

$$N_{\mu}=\left[\sum_{i\alpha} | S^{\mu}_{\mu i\alpha} | \right]^{-1}$$

to ensure that all of the order parameters tend to 1 when the spins are fully aligned in appropriate directions.

The first three order parameters are simply components of the uniform magnetization. We shall find it convenient to abbreviate these by writing M_{μ}, since there is here no ambiguity with the other (non-Ising) order parameters $M_{1\mu}$ and $M_{3\mu}$. η describes the well known six sublattice antiferromagnetic state in zero field,[14,15] and γ_{x^2} and γ_{y^2} the possible changes in the antiferromagnetic state which may be induced by a magnetic field.[16] The corresponding transformation properties are $M_{\mu}(T_1)$, $\eta(A_2)$, and $\gamma_{x^2},\gamma_{y^2}(E)$, where T_1, A_2, and E denote the usual irreducible representations of the cubic point group.

We shall find it convenient to express our results in terms of linear combinations of the three antiferromagnetic order parameters and we define

$$\eta_{\mu}=\sum_{i\alpha} \alpha S^{\mu}_{\mu i\alpha}N_{\mu} , \tag{1e}$$

so that

$$\eta=\tfrac{1}{3}(\eta_x+\eta_y+\eta_z) , \tag{1f}$$

$$\gamma_{x^2}=\tfrac{1}{2}(\eta_z-\eta_y) , \tag{1g}$$

$$\gamma_{y^2}=\tfrac{1}{2}(\eta_x-\eta_z) . \tag{1h}$$

The advantage of using the η_{μ} has been illustrated by Domann and Wolf[17] in connection with a model calculation of the effect of a magnetic field on the antiferromagnetic state. For symmetry directions for which $\eta_x=\eta_y$, it is sometimes convenient to use the single symbol η_{xy}.

B. Elastic strain

There are six independent components of the elastic strain ϵ_{pq} where $p,q=x,y,z$ and $\epsilon_{pq}=\epsilon_{qp}$. In a cubic material these may be grouped according to their transformation properties,

$$\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz} \quad (A_1) , \tag{2a}$$

$$\epsilon_{xx}-\epsilon_{yy},2\epsilon_{zz}-\epsilon_{xx}-\epsilon_{yy} \quad (E) , \tag{2b}$$

$$\epsilon_{xy},\epsilon_{yz},\epsilon_{zx} \quad (T_2) . \tag{2c}$$

As in the case of the magnetic order parameters, we shall sometimes find it convenient to express our results not in terms of these linear combinations, but in terms of the individual ϵ_{pq}. However, the symmetrized groupings are most useful for finding the appropriate invariants.

There are three invariants involving products of two of the ϵ_{pq}, corresponding to the usual elastic energy of a cubic crystal,[18]

$$E_{\rm el}=\tfrac{1}{2}C_{11}(\epsilon^2_{xx}+\epsilon^2_{yy}+\epsilon^2_{zz})+C_{12}(\epsilon_{xx}\epsilon_{yy}+\epsilon_{yy}\epsilon_{zz}+\epsilon_{zz}\epsilon_{xx}) +\tfrac{1}{2}C_{44}(\epsilon^2_{xy}+\epsilon^2_{yz}+\epsilon^2_{zx}) . \tag{3}$$

To study the magnetoelastic properties, we must now construct similar invariants involving the magnetic order parameters and the magnetic field.

C. Magnetoelastic invariants

We first consider terms linear in the strains and quadratic in the magnetic order parameters. (We can exclude terms linear in the magnetic order parameters by time reversal invariance.) Following standard group theoretical methods,[19] we find that there are nine invariants of this type, and these are listed in Table I. Similarly, we find that there are 28 invariants involving products of two strain components and two magnetic order parameters. These are listed in Table II.

In constructing these tables, it is convenient to make use of the transformation properties of both the magnetic order parameters and the elastic strains to check the number of invariants of a given type. For each case, we decompose the various reducible products into the corresponding irreducible representations, and count the number of occurrences of the identity representation. Thus, for example, if we look for terms containing products of two M's, we must decompose $(T_1\times T_1)_s=A_1+E+T_2$, where the subscript s denotes a symmetric product. Since the components of ϵ transform likewise as A_1+E+T_2, we see immediately that there will be just three invariants which are linear in the ϵ's and quadratic in the M's, as shown explicitly in Table I. Similarly, for terms quadratic in the ϵ's, we must decompose

$$[(A_1+E+T_2)\times(A_1+E+T_2)]_s =3A_1+3E+T_1+3T_2 .$$

Combining this with the decomposition of two M's, we see that there will be $3+3+3=9$ invariants quadratic in the ϵ's and quadratic in the M's, again as shown explicit-

TABLE I. Terms quadratic in the order parameters and linear in elastic strain.

$A_1\eta^2(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$
$A_2(\eta_x^2+\eta_y^2+\eta_z^2-\eta_y\eta_z-\eta_z\eta_x-\eta_x\eta_y)(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$
$A_3(M_x^2+M_y^2+M_z^2)(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$
$A_4(M_x^2\epsilon_{xx}+M_y^2\epsilon_{yy}+M_z^2\epsilon_{zz})$
$A_5\eta[(2\eta_x-\eta_y-\eta_z)\epsilon_{xx}+(2\eta_y-\eta_z-\eta_x)\epsilon_{yy}+(2\eta_z-\eta_x-\eta_y)\epsilon_{zz}]$
$A_6[(2\eta_x^2-\eta_y^2-\eta_z^2)\epsilon_{xx}+(2\eta_y^2-\eta_z^2-\eta_x^2)\epsilon_{yy}+(2\eta_z^2-\eta_x^2-\eta_y^2)\epsilon_{zz}]$
$A_7(M_yM_z\epsilon_{yz}+M_zM_x\epsilon_{zx}+M_xM_y\epsilon_{xy})$
$A_8[M_x(2\eta_x-\eta_y-\eta_z)\epsilon_{yz}+M_y(2\eta_y-\eta_z-\eta_x)\epsilon_{zx}+M_z(2\eta_z-\eta_x-\eta_y)\epsilon_{xy}]$
$A_9(M_x\epsilon_{yz}+M_y\epsilon_{zx}+M_z\epsilon_{xy})$

ly in Table II.

Whenever, as in the case of the terms quadratic in the ϵ's, a particular irreducible representation occurs more than once in the decomposition, there will be a corresponding arbitrariness in the definition of the invariants, since any linear combination will also be an invariant. In constructing Table II, we have tried to choose forms which will be as compact as possible, although this has sometimes resulted in expressions which do not reflect directly the invariants in the magnetic and elastic variables individually. We have also chosen to use the three η_μ, even though they do not transform as a single irre-

TABLE II. Terms quadratic in the order parameters and quadratic in elastic strain.

$B_1\eta^2(\epsilon_{xx}^2+\epsilon_{yy}^2+\epsilon_{zz}^2)$
$B_2(\eta_x^2+\eta_y^2+\eta_z^2-\eta_y\eta_z-\eta_z\eta_x-\eta_x\eta_y)(\epsilon_{xx}^2+\epsilon_{yy}^2+\epsilon_{zz}^2)$
$B_3(M_x^2+M_y^2+M_z^2)(\epsilon_{xx}^2+\epsilon_{yy}^2+\epsilon_{zz}^2)$
$B_4\eta^2(\epsilon_{yy}\epsilon_{zz}+\epsilon_{zz}\epsilon_{xx}+\epsilon_{xx}\epsilon_{yy})$
$B_5(\eta_x^2+\eta_y^2+\eta_z^2-\eta_y\eta_z-\eta_z\eta_x-\eta_x\eta_y)(\epsilon_{yy}\epsilon_{zz}+\epsilon_{zz}\epsilon_{xx}+\epsilon_{xx}\epsilon_{yy})$
$B_6(M_x^2+M_y^2+M_z^2)(\epsilon_{yy}\epsilon_{zz}+\epsilon_{zz}\epsilon_{xx}+\epsilon_{xx}\epsilon_{yy})$
$B_7\eta^2(\epsilon_{yz}^2+\epsilon_{zx}^2+\epsilon_{xy}^2)$
$B_8(\eta_x^2+\eta_y^2+\eta_z^2-\eta_y\eta_z-\eta_z\eta_x-\eta_x\eta_y)(\epsilon_{yz}^2+\epsilon_{zx}^2+\epsilon_{xy}^2)$
$B_9(M_x^2+M_y^2+M_z^2)(\epsilon_{yz}^2+\epsilon_{zx}^2+\epsilon_{xy}^2)$
$B_{10}\eta[(2\eta_x-\eta_y-\eta_z)\epsilon_{xx}^2+(2\eta_y-\eta_z-\eta_x)\epsilon_{yy}^2+(2\eta_z-\eta_x-\eta_y)\epsilon_{zz}^2]$
$B_{11}[(2\eta_x^2-\eta_y^2-\eta_z^2)\epsilon_{xx}^2+(2\eta_y^2-\eta_z^2-\eta_x^2)\epsilon_{yy}^2+(2\eta_z^2-\eta_x^2-\eta_y^2)\epsilon_{zz}^2]$
$B_{12}(M_x^2\epsilon_{xx}^2+M_y\epsilon_{yy}^2+M_z^2\epsilon_{zz}^2)$
$B_{13}\eta[(2\eta_x-\eta_y-\eta_z)\epsilon_{yy}\epsilon_{zz}+(2\eta_y-\eta_z-\eta_x)\epsilon_{zz}\epsilon_{xx}+(2\eta_z-\eta_x-\eta_y)\epsilon_{xx}\epsilon_{yy}]$
$B_{14}[(2\eta_x^2-\eta_y^2-\eta_z^2)\epsilon_{yy}\epsilon_{zz}+(2\eta_y^2-\eta_z^2-\eta_x^2)\epsilon_{zz}\epsilon_{xx}+(2\eta_z^2-\eta_x^2-\eta_y^2)\epsilon_{xx}\epsilon_{yy}]$
$B_{15}(M_x^2\epsilon_{yy}\epsilon_{zz}+M_y^2\epsilon_{zz}\epsilon_{xx}+M_z^2\epsilon_{xx}\epsilon_{yy})$
$B_{16}\eta[(2\eta_x-\eta_y-\eta_z)\epsilon_{yz}^2+(2\eta_y-\eta_z-\eta_x)\epsilon_{zx}^2+(2\eta_z-\eta_x-\eta_y)\epsilon_{xy}^2]$
$B_{17}[(2\eta_x^2-\eta_y^2-\eta_z^2)\epsilon_{yz}^2+(2\eta_y^2-\eta_z^2-\eta_x^2)\epsilon_{zx}^2+(2\eta_z^2-\eta_x^2-\eta_y^2)\epsilon_{xy}^2]$
$B_{18}(M_x^2\epsilon_{yz}^2+M_y^2\epsilon_{zx}^2+M_z^2\epsilon_{xy}^2)$
$B_{19}(M_yM_z\epsilon_{yz}+M_zM_x\epsilon_{zx}+M_xM_y\epsilon_{xy})(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$
$B_{20}(M_yM_z\epsilon_{xx}\epsilon_{yz}+M_zM_x\epsilon_{yy}\epsilon_{zx}+M_xM_y\epsilon_{zz}\epsilon_{xy})$
$B_{21}\eta(M_x\epsilon_{yz}+M_y\epsilon_{zx}+M_z\epsilon_{xy})(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$
$B_{22}\eta(M_x\epsilon_{xx}\epsilon_{yz}+M_y\epsilon_{yy}\epsilon_{zx}+M_z\epsilon_{zz}\epsilon_{xy})$
$B_{23}[M_x(2\eta_x-\eta_y-\eta_z)\epsilon_{yz}+M_y(2\eta_y-\eta_z-\eta_x)\epsilon_{zx}+M_z(2\eta_z-\eta_x-\eta_y)\epsilon_{xy}](\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$
$B_{24}[M_x(2\eta_x-\eta_y-\eta_z)\epsilon_{xx}\epsilon_{yz}+M_y(2\eta_y-\eta_z-\eta_x)\epsilon_{yy}\epsilon_{zx}+M_z(2\eta_z-\eta_x-\eta_y)\epsilon_{zz}\epsilon_{xy}]$
$B_{25}[M_x(\eta_y-\eta_z)(\epsilon_{yy}-\epsilon_{zz})\epsilon_{yz}+M_y(\eta_z-\eta_x)(\epsilon_{zz}-\epsilon_{xx})\epsilon_{zx}+M_z(\eta_x-\eta_y)(\epsilon_{xx}-\epsilon_{yy})\epsilon_{xy}]$
$B_{26}(M_yM_z\epsilon_{xy}\epsilon_{zx}+M_zM_x\epsilon_{yz}\epsilon_{xy}+M_xM_y\epsilon_{zx}\epsilon_{yz})$
$B_{27}\eta(M_x\epsilon_{xy}\epsilon_{zx}+M_y\epsilon_{yz}\epsilon_{xy}+M_z\epsilon_{zx}\epsilon_{yz})$
$B_{28}[M_x(2\eta_x-\eta_y-\eta_z)\epsilon_{xy}\epsilon_{zx}+M_y(2\eta_y-\eta_z-\eta_x)\epsilon_{yz}\epsilon_{xy}+M_z(2\eta_z-\eta_x-\eta_y)\epsilon_{zx}\epsilon_{yz}]$

TABLE III. Terms linear in the order parameters, linear in field, and linear inelastic strain.

$$a_3(M_xH_x+M_yH_y+M_zH_z)(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$$
$$a_4(M_xH_x\epsilon_{xx}+M_yH_y\epsilon_{yy}+M_zH_z\epsilon_{zz})$$
$$a_7[\tfrac{1}{2}(M_yH_z+M_zH_y)\epsilon_{yz}+\tfrac{1}{2}(M_zH_x+M_xH_z)\epsilon_{zx}+\tfrac{1}{2}(M_xH_y+M_yH_x)\epsilon_{xy}]$$
$$a_8[H_x(2\eta_x-\eta_y-\eta_z)\epsilon_{yz}+H_y(2\eta_y-\eta_z-\eta_x)\epsilon_{zx}+H_z(2\eta_z-\eta_x-\eta_y)\epsilon_{xy}]$$
$$a_9\eta(H_x\epsilon_{yz}+H_y\epsilon_{zx}+H_z\epsilon_{xy})$$

ducible representation. If one uses instead the proper cubic variables η, γ_{x^2}, and γ_{y^2}, as defined in Ref. 16, one tends to lose the evident symmetry under the cyclic permutation of x, y, and z, corresponding to the threefold symmetry operation, and the simplifications which can arise when $\eta_x=\eta_y=\eta_z$. Of course, the number of *independent* invariants is not affected by these choices, and we must always end up with a nine linear and 28 quadratic terms in the ϵ's, as indicated by the general considerations discussed above.

If we were using the standard Landau theory approach, in which all of the order parameters are considered to be small, the terms shown in Tables I and II would give the complete expression needed to discuss magnetoelastic effects. In fact, the order parameters are generally not small and they vary widely with field and temperature, as illustrated explicitly by the model calculation of Domann and Wolf.[17] One must inquire, therefore, under what conditions the terms in Tables I and II may provide a useful first approximation for the magnetoelastic energy, or if one may need additional terms.

If one considers the possible physical origins for magnetoelastic effects in a system such as DyAlG, it is clear that one major contribution will come from the strain dependence of the microscopic spin-spin interaction. In a mean-field approximation, the spin-spin interaction can be expressed in terms of products of just pairs of order parameters, one representing the mean field which acts on the other. In this approximation, the terms given in Tables I and II are therefore complete and no higher powers of the magnetic order parameters need to be considered.

However, there is a second mechanism which is not covered by this approximation and which will lead to some important terms which are not usually considered. This mechanism involves the strain dependence of the crystal field which acts on the Dy^{3+} ions. The result of this strain dependence can be thought of as a variation of the magnetic g values. The importance of this mechanism for DyAlG was noted by Dillon *et al.*[6] but no explicit calculations were reported. We will discuss this mechanism in more detail in Sec. IV, but for now we simply note that it will introduce additional terms which are linear in the magnetic field and linear in one of the six order parameters.

We can readily find the corresponding terms in the magnetoelastic energy from the terms we have already derived in Tables I and II, by simply replacing one component of **M** by the corresponding component of the applied field **H**. This is evident from the fact that the components of both **M** and **H** transform as T_1. The resulting terms are listed in Tables III and IV.

We see, therefore, that the magnetoelastic energy for DyAlG contains no less than 58 independent invariants, even in this simple approximation. This is a formidable number, but we shall see in the next section that many terms will be zero for fields applied along symmetry directions, and in the limiting cases of high and low

TABLE IV. Terms linear in the order parameters, linear in field, and quadratic in elastic strain.

$$b_3(M_xH_x+M_yH_y+M_zH_z)(\epsilon_{xx}^2+\epsilon_{yy}^2+\epsilon_{zz}^2)$$
$$b_6(M_xH_x+M_yH_y+M_zH_z)(\epsilon_{yy}\epsilon_{zz}+\epsilon_{zz}\epsilon_{xx}+\epsilon_{xx}\epsilon_{yy})$$
$$b_9(M_xH_x+M_yH_y+M_zH_z)(\epsilon_{yz}^2+\epsilon_{zx}^2+\epsilon_{xy}^2)$$
$$b_{12}(M_xH_x\epsilon_{xx}^2+M_yH_y\epsilon_{yy}^2+M_zH_z\epsilon_{zz}^2)$$
$$b_{15}(M_xH_x\epsilon_{yy}\epsilon_{zz}+M_yH_y\epsilon_{zz}\epsilon_{xx}+M_zH_z\epsilon_{xx}\epsilon_{yy})$$
$$b_{18}(M_xH_x\epsilon_{yz}^2+M_yH_y\epsilon_{zx}^2+M_zH_z\epsilon_{xy}^2)$$
$$b_{12}[\tfrac{1}{2}(M_yH_z+M_zH_y)\epsilon_{yz}+\tfrac{1}{2}(M_zH_x+M_xH_z)\epsilon_{zx}+\tfrac{1}{2}(M_xH_y+M_yH_x)\epsilon_{xy}](\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$$
$$b_{20}[\tfrac{1}{2}(M_yH_z+M_zH_y)\epsilon_{xx}\epsilon_{yz}+\tfrac{1}{2}(M_zH_x+M_xH_z)\epsilon_{yy}\epsilon_{xz}+\tfrac{1}{2}(M_xH_y+M_yH_x)\epsilon_{zz}\epsilon_{xy}]$$
$$b_{21}(H_x\epsilon_{yz}+H_y\epsilon_{zx}+H_z\epsilon_{xy})(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$$
$$b_{22}\eta(H_x\epsilon_{xx}\epsilon_{yz}+H_y\epsilon_{yy}\epsilon_{zx}+H_z\epsilon_{zz}\epsilon_{xy})$$
$$b_{23}[H_x(2\eta_x-\eta_y-\eta_z)\epsilon_{yz}+H_y(2\eta_y-\eta_z-\eta_x)\epsilon_{zx}+H_z(2\eta_z-\eta_x-\eta_y)\epsilon_{xy}](\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$$
$$b_{24}[H_x(2\eta_x-\eta_y-\eta_z)\epsilon_{xx}\epsilon_{yz}+H_y(2\eta_y-\eta_z-\eta_x)\epsilon_{yy}\epsilon_{zx}+H_z(2\eta_z-\eta_x-\eta_y)\epsilon_{zz}\epsilon_{xy}]$$
$$b_{25}[H_x(\eta_y-\eta_z)(\epsilon_{yy}-\epsilon_{zz})\epsilon_{yz}+H_y(\eta_z-\eta_x)(\epsilon_{zz}-\epsilon_{xx})\epsilon_{zx}+H_z(\eta_x-\eta_y)(\epsilon_{xx}-\epsilon_{yy})\epsilon_{xy}]$$
$$b_{26}[\tfrac{1}{2}(M_yH_z+M_zH_y)\epsilon_{xy}\epsilon_{zx}+\tfrac{1}{2}(M_zH_x+M_xH_z)\epsilon_{yz}\epsilon_{xy}+\tfrac{1}{2}(M_xH_y+M_yH_x)\epsilon_{zx}\epsilon_{yz}]$$
$$b_{27}(H_x\epsilon_{xy}\epsilon_{zx}+H_y\epsilon_{yz}\epsilon_{xy}+H_z\epsilon_{zx}\epsilon_{yz})$$
$$b_{28}[H_x(2\eta_x-\eta_y-\eta_z)\epsilon_{xy}\epsilon_{zx}+H_y(2\eta_y-\eta_z-\eta_x)\epsilon_{yz}\epsilon_{xy}+H_z(2\eta_z-\eta_x-\eta_y)\epsilon_{zx}\epsilon_{yz}]$$

fields. In some cases, only one or two terms are, in fact, required.

More important is the consideration of the validity of the approximation which we have used. As stated above, it is effectively a mean-field based approximation and, as such, it is clearly not very good near phase transitions. However, for high fields or for low temperatures, such as approximation becomes asymptotically *exact,* since the excitations in an Ising-like system such as DyAlG are simple spin flips and not spin waves. In other regions, we can only expect the theory to be semiquantitative.

We should also note that we have neglected a large number of additional terms, which are allowed by symmetry, when we limit the order parameters to the six corresponding to the Ising approximation. In fact, DyAlG is not *exactly* an Ising model, and 12 additional order parameters may be nonzero.[16] These can, of course, also couple both to the other order parameters and to the field through the strain. We shall not discuss these terms here as their effect would appear to be small for the particular observations which we shall be considering, but it is important to keep in mind that such terms do exist and that they may be important in some circumstances.

D. Magnetostriction

Combining the linear magnetoelastic terms (Table I and III) with the purely elastic energy [Eq. (3)] and differentiating in turn with respect to the various strain components, we can find the magnetoelastic strain for any given field and set of magnetic order parameters. We shall consider two specific examples in Sec. III A. The individual strain components must then be related to the actual changes of length (Δl) measured in a magnetostriction experiment. In Table V, we list a number of these relationships for magnetostriction measured along the principal directions in a cubic crystal. This table is derived simply from the definitions of the strain components[8] but it is useful for considering different cases.

E. Magnetoacoustic effects

We can identify the various components of the magnetoelastic tensor, ΔC_{ijkl}, by picking out all of the coefficients of a given product $\epsilon_{ij}\epsilon_{kl}$. Thus, for example, the coefficient of ϵ_{zz}^2 is seen to be

TABLE V. Magnetostriction along the principal directions.

H‖[111]

$$\left[\frac{\Delta l}{l}\right]_{[111]} = -\frac{P}{C_{11}+2C_{12}} - \frac{2Q}{C_{44}}$$

$$\left[\frac{\Delta l}{l}\right]_{[1\bar{1}0]} = -\frac{P}{C_{11}+2C_{12}} + \frac{Q}{C_{44}},$$

Definitions

$$P = A_1\eta^2 + (A_3 + \tfrac{1}{3}A_4)M^2 + (a_3 + \tfrac{1}{3}a_4)MH,$$

$$Q = \tfrac{1}{3}A_7M^2 + \frac{1}{\sqrt{3}}A_9\eta M + \tfrac{1}{3}a_7MH + \frac{1}{\sqrt{3}}a_9\eta H.$$

H‖[001]

$$\left[\frac{\Delta l}{l}\right]_{[001]} = -\frac{(C_{11}+C_{12})Q - 2C_{12}P}{(C_{11}-C_{12})(C_{11}+2C_{12})}$$

$$\left[\frac{\Delta l}{l}\right]_{[110]} = -\frac{C_{11}P - C_{12}Q}{(C_{11}-C_{12})(C_{11}+2C_{12})} - \frac{S}{C_{44}}$$

$$\left[\frac{\Delta l}{l}\right]_{[1\bar{1}0]} = -\frac{C_{11}P - C_{12}Q}{(C_{11}-C_{12})(C_{11}+2C_{12})} - \frac{S}{C_{44}},$$

Definitions

$$P = A_1\eta^2 + A_2\Delta\eta^2 + A_3M^2 + A_5\eta\Delta\eta + A_6(\eta_{xy}^2 - \eta_z^2) + a_3MH,$$

$$Q = A_1\eta^2 + A_2\Delta\eta^2 + (A_3 + A_4)M^2 - 2A_5\eta\Delta\eta - 2A_5\eta\Delta - 2A_6(\eta_{xy}^2 - \eta_z^2) + (a_3 + a_4)MH,$$

$$S = -2A_8M\Delta\eta + A_9\eta M - 2a_8\Delta\eta H + a_9\eta_H.$$

TABLE V. (*Continued*)

H‖[110]

$$\left[\frac{\Delta l}{l}\right]_{[110]} = -\frac{C_{11}P - C_{12}Q}{(C_{11}-C_{12})(C_{11}+2C_{12})} - \frac{S}{C_{44}}$$

$$\left[\frac{\Delta l}{l}\right]_{[\bar{1}10]} = -\frac{C_{11}P - C_{12}Q}{(C_{11}-C_{12})(C_{11}+2C_{12})} + \frac{S}{C_{44}}$$

$$\left[\frac{\Delta l}{l}\right]_{[001]} = -\frac{(C_{11}+C_{12})Q - 2C_{12}P}{(C_{11}-C_{12})(C_{11}+2C_{12})}$$

$$\left[\frac{\Delta l}{l}\right]_{[\bar{1}11]} = -\tfrac{1}{3}\frac{2P+Q}{C_{11}+2C_{12}} - \frac{2}{3}\frac{S}{C_{44}},$$

Definitions

$$P = A_1\eta^2 + A_2\Delta\eta^2 + (A_3 + \tfrac{1}{2}A_4)M^2 + A_5\eta\Delta\eta + A_6(\eta_{xy}^2 - \eta_z^2) + (a_3 + \tfrac{1}{2}a_4)MH,$$

$$Q = A_1\eta^2 + A_2\Delta\eta^2 + A_3M^2 - 2A_5\eta\Delta\eta - 2A_6(\eta_{xy}^2 - \eta_z^2) + a_3MH,$$

$$S = \tfrac{1}{2}A_7M^2 + \tfrac{1}{2}a_7MH.$$

$$\tfrac{1}{2}\Delta C_{zzzz} = B_1\eta^2 + B_2(\eta_x^2 + \eta_y^2 + \eta_z^2 - \eta_x\eta_y - \eta_y\eta_z - \eta_z\eta_x) + B_3(M_x^2 + M_y^2 + M_z^2) + B_{10}\eta(2\eta_z - \eta_x - \eta_y)$$
$$+ B_{11}(2\eta_z^2 - \eta_x^2 - \eta_y^2) + B_{12}M_z^2 + b_3(M_xH_x + M_yH_y + M_zH_z) + b_{12}M_zH_z\ , \tag{4}$$

with similar expressions for the other components. For a given magnetic state, specified in terms of the M's and η's and the field, we can thus find the ΔC's in terms of the coefficients B_n and b_n. We shall find it convenient to use the standard notation for elastic tensors[18] in which $xx=1$, $yy=2$, $zz=3$, $yz=4$, $zx=5$, and $xy=6$, so that $\Delta C_{zzzz} = \Delta C_{33}$. The results for the relevant ΔC's are shown in Table VI.

The ΔC's are closely related to changes in velocity Δv of acoustic waves propagating through the material, which are generally used to study magnetoelastic effects. We can use standard expressions relating ΔC to Δv,[20] but there two special problems to which we must pay attention.

One is the fact that the symmetry in the magnetic state may be lower than one might perhaps expect, so that the identification of the normal modes may not always be obvious. For example, for a field applied along [001] one might have expected the symmetry to be tetragonal, starting from cubic in zero field, so that the velocity of transverse waves propagating along [001] should be independent of polarization. This is true in the paramagnetic state, but not, in fact, when $\eta \neq 0$. We can see this by noting the two nonzero magnetostrictive terms of the form $A_9\epsilon_{xy}\eta M_z$ and $a_9\epsilon_{xy}\eta H_z$ (see Tables I and III), which show that a field along [001] will produce a distortion along [110], which is positive or negative according to the sign of the antiferromagnetic order parameter η. The resulting symmetry is thus *orthorhombic*, with principal axes along [110] and [1$\bar{1}$0] and [001], and these will also be the axes for the normal modes. The corresponding term in the magnetoelastic tensor is ΔC_{45}, which would be absent if the symmetry were tetragonal. Similarly, for **H**‖[110] the symmetry is only monoclinic, with [1$\bar{1}$0] as the unique axis. For **H**‖[111] the symmetry is trigonal, as one might expect naively.

The lower symmetry results in the fact that the crystal can be acoustically *birefringent*, a finding which is at first surprising for a material which is basically cubic. Of course the birefringence is small, but so is the entire magnetoelastic effect.

In practice, the change of symmetry demands a change of axes of the magnetoelastic energy expression from the original cubic axes used in Tables II and IV. For the particular cases which we shall consider, the changes are quite simple. For H‖[001] and H‖[110], a rotation of $\pi/4$ about the appropriate symmetry axis is required. For H‖[111], a rotation of $\pi/4$ about the [001] axis followed by a rotation of $\cos^{-1}(1/\sqrt{3})$ about the original [1$\bar{1}$0] axis makes the z axis coincide with [111]. A convenient method for transforming the elastic constants is given in Chap 3 of Ref. 20.

The second problem, which is unusual, is the fact that magnetostrictive effects will change ρ, the *density* of the material, so that the velocity, which is given by $\sqrt{C/\rho}$, will be affected not only by ΔC but also by $\Delta\rho$. Thus

$$\frac{\Delta v}{v} = \frac{1}{2}\frac{\Delta C}{C} - \frac{1}{2}\frac{\Delta\rho}{\rho}\ , \tag{5}$$

where $\Delta\rho/\rho$ is related to the volume expansion $\tilde{V} = \Delta V/V$ by

$$\frac{\Delta\rho}{\rho}=-\tilde{V}=-\frac{\Delta V}{V}\ . \tag{6}$$

This effect is, presumably, present in all magnetoelastic experiments using acoustic velocity measurements, but it is generally neglected since $\tilde{V}$ is usually small relative to the change in velocity.[21] However, when, as in our case, $\Delta C/C$ itself is small, the effect may not be negligible.[22]

We shall defer discussion of the correction until we examine specific examples. Here we shall concentrate on deriving the appropriate expressions for $\Delta v/v$ in terms of the ΔC's for some of the propagation modes which may be useful for studying the magnetoelastic effects. We can make use of the slowness curves listed in Appendix 3B of Ref. 20., and in Table VI we summarize the expressions which give $\Delta v/v$ for various modes in the absence of the magnetostrictive corrections. Together with Tables II and IV one can now find $\Delta v/v$ as a function of the M's and η's and the applied field. We shall illustrate the procedure in Sec. III B.

TABLE VI. Magnetoelastic contributions to velocity changes.

H∥[111]

Conditions	Contributions
$\underline{k}\parallel[111]$, $\epsilon\parallel[111]$	$\frac{\Delta v}{v}=\frac{1}{2}\frac{\Delta C_{11}+2\Delta C_{12}+4\Delta C_{14}+8\Delta C_{15}+4\Delta C_{44}+8\Delta C_{45}}{C_{11}+2C_{12}+4C_{44}}$
$\underline{k}\parallel[111]$, $\epsilon\perp[111]$	$\frac{\Delta v}{v}=\frac{1}{2}\frac{\Delta C_{11}-\Delta C_{12}+\Delta C_{44}-\Delta C_{45}-2\Delta C_{14}+2\Delta C_{15}}{C_{11}-C_{12}+C_{44}}$

Definitions

$$\tfrac{1}{2}\Delta C_{11}=\tfrac{1}{2}\Delta C_{22}=\tfrac{1}{2}\Delta C_{33}=B_1\eta^2+(B_3+\tfrac{1}{3}B_{12})M^2+(b_3+\tfrac{1}{3}b_{12})MH$$

$$\tfrac{1}{2}\Delta C_{44}=\tfrac{1}{2}\Delta C_{55}=\tfrac{1}{2}\Delta C_{66}=B_7\eta^2+(B_9+\tfrac{1}{3}B_{18})M^2+(b_9+\tfrac{1}{3}b_{18})MH$$

$$\Delta C_{12}=\Delta C_{13}=\Delta C_{23}=B_4\eta^2+(B_6+\tfrac{1}{3}B_{15})M^2+(b_6+\tfrac{1}{3}b_{15})MH$$

$$\Delta C_{14}=\Delta C_{25}=\Delta C_{36}=\tfrac{1}{3}(B_{19}+B_{20})M^2+\frac{1}{\sqrt{3}}(B_{21}+B_{22})M\eta+\tfrac{1}{3}(b_{19}+b_{20})MH+\frac{1}{\sqrt{3}}(b_{21}+b_{22})\eta H$$

$$\Delta C_{15}=\Delta C_{16}=\Delta C_{24}=\Delta C_{26}=\Delta C_{34}=\Delta C_{35}$$
$$=\tfrac{1}{3}B_{19}M^2+\frac{1}{\sqrt{3}}B_{21}M\eta+\tfrac{1}{3}b_{19}MH+\frac{1}{\sqrt{3}}b_{21}\eta H$$

$$\Delta C_{45}=\Delta C_{46}=\Delta C_{56}$$
$$=\tfrac{1}{3}B_{26}M^2+\frac{1}{\sqrt{3}}B_{27}M\eta+\tfrac{1}{3}b_{26}MH+\frac{1}{\sqrt{3}}b_{27}\eta H$$

H∥[001]

Conditions	Contributions
$\underline{k}\parallel[001]$, $\epsilon\parallel[001]$	$\frac{\Delta v}{v}=\frac{\Delta C_{23}}{2C_{11}}$
$\underline{k}\parallel[001]$, $\epsilon\parallel[110]$	$\frac{\Delta v}{v}=\frac{\Delta C_{44}+\Delta C_{45}}{2C_{44}}$
$\underline{k}\parallel[001]$, $\epsilon\parallel[1\bar{1}0]$	$\frac{\Delta v}{v}=\frac{\Delta C_{44}-\Delta C_{45}}{2C_{44}}$
$\underline{k}\parallel[110]$, $\epsilon\parallel[110]$	$\frac{\Delta v}{v}=\frac{1}{2}\frac{\Delta C_{11}+\Delta C_{12}+4\Delta C_{16}+2\Delta C_{66}}{C_{11}+C_{12}+2C_{44}}$
$\underline{k}\parallel[1\bar{1}0]$, $\epsilon\parallel[1\bar{1}0]$	$\frac{\Delta v}{v}=\frac{1}{2}\frac{\Delta C_{11}+\Delta C_{12}-4\Delta C_{16}+2\Delta C_{66}}{C_{11}+C_{12}+2C_{44}}$
$\underline{k}\parallel[110]$, $\epsilon\parallel[1\bar{1}0]$	$\frac{\Delta v}{v}=\frac{1}{2}\frac{\Delta C_{11}-\Delta C_{12}}{C_{11}-C_{12}}$

Definitions

$$\tfrac{1}{2}\Delta C_{11}=\tfrac{1}{2}\Delta C_{22}=B_1\eta^2+B_2(\Delta\eta)^2+B_3M^2+B_{10}\eta\Delta\eta+B_{11}(\eta_{xy}^2-\eta_z^2)+b_3MH$$

$$\tfrac{1}{2}\Delta C_{33}=B_1\eta^2+B_2(\Delta\eta)^2+(B_3+B_{12})M^2-2B_{10}\eta\Delta\eta-2B_{11}(\eta_{xy}^2-\eta_z^2)+(b_3+b_{12})MH$$

$$\tfrac{1}{2}\Delta C_{44}=\tfrac{1}{2}\Delta C_{55}=B_7\eta^2+B_8(\Delta\eta)^2+B_9M^2+B_{16}\eta\Delta\eta+B_{17}(\eta_{xy}^2-\eta_z^2)+b_9MH$$

$$\tfrac{1}{2}\Delta C_{66}=B_7\eta^2+B_8(\Delta\eta)^2+(B_9+B_{18})M^2-2B_{16}\eta\Delta\eta-2B_{17}(\eta_{xy}^2-\eta_z^2)+(b_9+b_{18})MH$$

$$\Delta C_{12}=B_4\eta^2+B_5(\Delta\eta)^2+(B_6+B_{15})M^2-2B_{13}\eta\Delta\eta-2B_{14}(\eta_{xy}^2-\eta_z^2)+(b_6+b_{15})MH$$

$$\Delta C_{16}=\Delta C_{26}=B_{21}M\Delta\eta-2B_{23}M\Delta\eta+b_{21}\eta H-2b_{23}\Delta\eta H$$

$$\Delta C_{45}=B_{27}M\eta-2B_{28}M\Delta\eta+b_{27}\eta H-2b_{28}\Delta\eta H$$

TABLE VI. (*Continued*)

H‖[110]	
Conditions	Contributions
$\underline{k}\|\|[1\bar{1}0]$, $\underline{\epsilon}\|\|[1\bar{1}0]$	$\frac{\Delta v}{v}=\frac{1}{2}\frac{\Delta C_{11}+\Delta C_{12}-4\Delta C_{16}+2\Delta C_{66}}{C_{11}+C_{12}+2C_{44}}$
$\underline{k}\|\|[110]$, $\underline{\epsilon}\|\|[1\bar{1}0]$	$\frac{\Delta v}{v}=\frac{1}{2}\frac{\Delta C_{11}-\Delta C_{12}}{C_{11}-C_{12}}$
$\underline{k}\|\|[001]$, $\underline{\epsilon}\|\|[1\bar{1}0]$	$\frac{\Delta v}{v}=\frac{1}{2}\frac{\Delta C_{44}-\Delta C_{45}}{\Delta C_{44}}$,

Definitions

$\frac{1}{2}\Delta C_{11}=\frac{1}{2}\Delta C_{22}=B_1\eta^2+B_2(\Delta\eta)^2+(B_3+\frac{1}{2}B_{12})M^2+B_{10}\eta\Delta\eta+B_{11}(\eta_{xy}^2-\eta_z^2)+(b_3+\frac{1}{2}b_{12})MH$

$\frac{1}{2}\Delta C_{44}=\frac{1}{2}C_{55}=B_7\eta^2+B_8(\Delta\eta)^2+(B_9+\frac{1}{2}B_{18})M^2+B_{16}\eta\Delta\eta+B_{17}(\eta_{xy}^2-\eta_z^2)+(b_9+\frac{1}{2}b_{18})MH$

$\frac{1}{2}\Delta C_{66}=B_7\eta^2+B_8(\Delta\eta)^2+B_9M^2-2B_{16}\eta\Delta\eta-2B_{17}(\eta_{xy}^2-\eta_z^2)+b_9MH,$

$\Delta C_{12}=B_4\eta^2+B_5(\Delta\eta)^2+B_6M^2-2B_{13}\eta\Delta\eta-2B_{14}(\eta_{xy}^2-\eta_z^2)+b_6MH,$

$\Delta C_{16}=\Delta C_{26}=\frac{1}{2}B_{19}M^2+\frac{1}{2}b_{19}MH$

$\Delta C_{45}=\frac{1}{2}B_{26}M^2+\frac{1}{2}b_{26}MH$

III. APPLICATIONS

In this section we shall apply the general theory developed above to a number of specific cases in which unusual magnetoelastic behavior has been observed. These examples will illustrate some of the many predictions which can be made..

A. Magnetostriction

1. *Field parallel to [111]*

The experimental results obtained by Dillon *et al.*[6] for this case are reproduced in Fig. 1. A number of striking features are immediately apparent.

(a) For low fields, the axial magnetostriction is linear in field, changing sign at $H=0$. The corresponding volume magnetostriction is very small and similar for positive and negative fields.

(b) For high fields, the axial magnetostriction is again approximately linear in field, with a slope that is independent of the sign of H and essentially independent of temperature. The corresponding volume magnetostriction, on the other hand, varies with temperature, tending to a constant value, which is independent of field.

To interpret these features, we first use the terms in Tables I and III together with Eq. (3) to find the magnetostrictive strains, by minimizing with respect to each of the ϵ_{pq} in turn. We find

$$\epsilon_{xx}=\epsilon_{yy}=\epsilon_{zz}=\frac{-A_1\eta^2-(A_3+A_4/3)M^2-(a_3+a_4/3)MH}{C_{11}+2C_{12}} \tag{7}$$

and

$$\epsilon_{xy}=\epsilon_{yz}=\epsilon_{zx}=\frac{-A_7M^2-\sqrt{3}A_9\eta M-2a_7MH-\sqrt{3}a_9\eta H}{3C_{44}}\ , \tag{8}$$

where M and H are the magnetization and field along [111].

Using Table V, we can then find expressions for the axial distortion $\tilde{E}$ as defined in Ref. 6.

$$\tilde{E}=\frac{\Delta l_{\|}}{l_{\|}}-\frac{\Delta l_{\perp}}{l_{\perp}}=\tfrac{3}{2}\epsilon_{xy}\ , \tag{9a}$$

and for the volume distortion $\tilde{V}$,

$$\tilde{V}=\frac{\Delta l_{\|}}{l_{\|}}+2\frac{\Delta l_{\perp}}{l_{\perp}}$$

$$=(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})_H-(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})_{H=0}\ . \tag{9b}$$

We see that $\tilde{E}$ has the form

$$\tilde{E}=f_1M^2+f_2\eta M+f_3\eta H+f_4MH\ , \tag{10}$$

while $\tilde{V}$ has the form

$$\tilde{V}=f_5\eta^2+f_6M^2+f_7MH\ , \tag{11}$$

where $f_1, f_2 \ldots$ etc., are parameters containing the A_n, a_n, and C_{ij}.

For low fields and low temperatures, these reduce to

$$\tilde{E}=f_3\eta H\ , \tag{10'}$$

$$\tilde{V}=f_5\eta^2\ , \tag{11'}$$

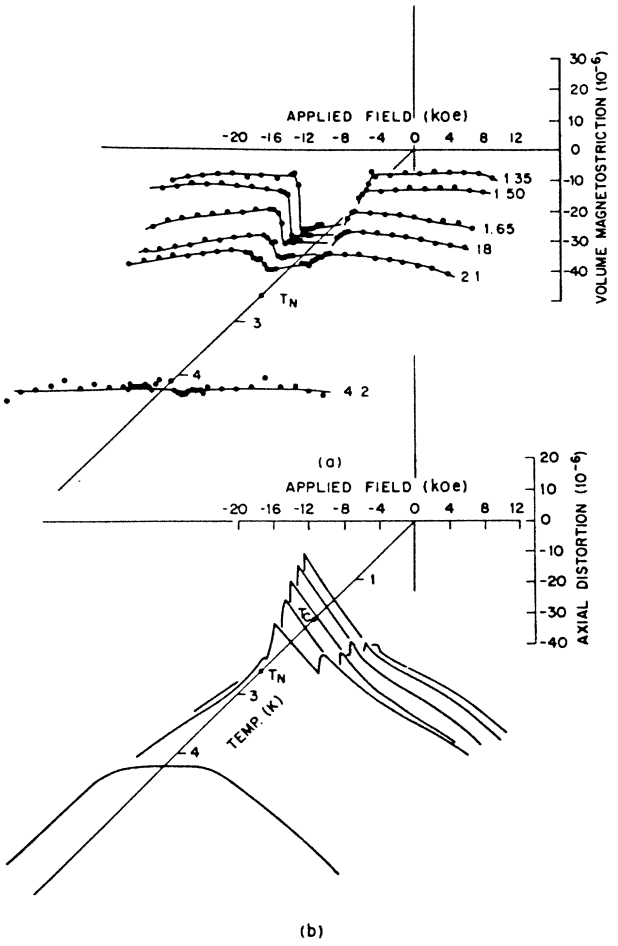

FIG. 1. (a) Volume magnetostriction, $\tilde{V}$, and (b) axial distortion, $\tilde{E}$, for DyAlG as a function of magnetic field applied parallel to [111] for various temperatures. After Ref. 6.

while for high fields and low temperatures, we find

$$\tilde{E}=f_1M^2+f_4MH\ , \tag{10''}$$

$$\tilde{V}=f_6M^2+f_7MH\ . \tag{11''}$$

From Eq. (10') we see immediately the origin of the finite slope at low fields which is observed for $\tilde{E}$, and from Eq. (11') we see the lack of dependence on field observed at low fields for $\tilde{V}$. Equation (11') also shows that $\tilde{V}$ should be dependent on temperature, varying with η^2 from zero at the Néel temperature to a constant value as $T\rightarrow 0$ K. This change was assumed to be negligible by Dillon *et al.* when they constructed Fig. 1, based on earlier thermal expansion measurements.[23] In the absence of additional data, we can only conclude either that f_5 is very small, or that the expansion measurements may somehow have underestimated the effect. We shall discuss this further below.

From the observed lack of field dependence of $\tilde{V}$ at low temperatures we must conclude that f_7 is small, while the lack of field dependence at 4.2 K show that f_5 is small also. On the other hand, the large field dependence of $\tilde{E}$ at high fields show that f_4 is large. The term f_4MH is also consistent with the similar behavior of $\tilde{E}$ in positive and negative fields and it also explains the lack of temperature dependence of the high-field slopes, since M will be essentially saturated at all of the temperatures studied for fields ~ 10 kOe (1 T).

On the basis of this analysis we can conclude that, effectively, only two or three terms dominate the magnetostrictive behavior for H||[111]. The two large terms are clearly those proportional to f_3 and f_4, which give rise to the strong field dependence of $\tilde{E}$. The observed jump in $\tilde{V}$ as a function of field can only be interpreted, in terms of the present theory, if the third parameter f_5 is not in fact small, since f_6 and f_7 clearly must be small from the behavior at 4.2 K. It would be very interesting to repeat the earlier thermal expansion measurements[23] to see if there was in fact some systematic error.

An independent indication that there is some inconsistency in the present experimental picture is provided by a comparison of the high-field values of $\tilde{V}$ at the lowest and highest temperatures. For fields of 12 kOe (1.2 T) along [111], DyAlG should be in a very similar state at 4.2 and 1.35 K, corresponding to essentially complete magnetic saturation. It is very difficult to see why the volume magnetostriction should be very different under these conditions. This paradox would be resolved if f_5 was in fact large, so that the curves in the antiferromagnetic state would be shifted down when $\eta\neq 0$.

The above analysis can be used to obtain semiquantitative estimates of the parameters f_3, f_4, and f_5 from the experimental data. These can then be related to the corresponding magnetoelastic parameters a_5, a_7, and A_1, if the elastic constants are also known.

This raises the matter of units. The simplest procedure is to define each of the order parameters as a dimensionless quantity which reaches its maximum value of 1 at $T=0$ K. The parameters A_n and B_n then have units of energy per unit volume, $\mathrm{J\,m^{-3}}$ in SI. We can choose the same units for the parameters a_n and b_n by measuring the field in tesla.

From the experimental data in Fig. 1, we can estimate

$$\tilde{E}=(38\times 10^{-6})H \tag{12}$$

for low fields and low temperatures, and

$$\tilde{E}=-(20\times 10^{-6})\,|\,H\,| \tag{13}$$

for high fields. The corresponding discontinuity in $\tilde{V}$ is estimated to be

$$\Delta\tilde{V}\approx 20\times 10^{-6}\ . \tag{14}$$

We also need values for the elastic constants for DyAlG. These have recently been estimated by Huan[24] using pulse echo measurements. He finds

$$\begin{aligned} C_{11}&=3.47\times 10^{11}\ \mathrm{J\,m^{-3}}\ ,\\ C_{12}&=1.00\times 10^{11}\ \mathrm{J\,m^{-3}}\ ,\\ C_{44}&=1.11\times 10^{11}\ \mathrm{J\,m^{-3}}\ , \end{aligned} \tag{15}$$

with an estimated accuracy of about ±6%. Using Eqs. (10) and (11) with the experimental results [Eqs. (12)–(15)], we then find

$$a_9 = -4.9\times10^6\ \mathrm{J\,m^{-3}\,T^{-1}}\ ,$$
$$a_7 = 2.6\times10^6\ \mathrm{J\,m^{-3}\,T^{-1}}\ , \tag{16}$$
$$A_1 = 3.7\times10^6\ \mathrm{J\,m^{-3}}\ .$$

The other parameters, A_3, A_4, A_7, a_3, and a_4, in Eqs. (7) and (8), are much smaller, and are not determinable from the present data. The parameter A_9 will be important only in regions where both M and η are large, i.e., below T_N but not too far below, since M is then small, but it will be hard to separate this term from the generally larger term in a_9. Qualitatively, the two give similar effects.

2. *Field parallel to [001]*

It is interesting to compare the previous results with those for the field applied along [001]. Measurements for this orientation have been carried out by Dillon, Chen, and Gyorgy,[13] and in Fig. 2 we reproduce their results for $(\Delta l_\parallel/l_\parallel)=(\Delta l/l)_{[001]}$ and $\Delta l_\perp/l_\perp=(\Delta l/l)_{[100]}$. A number of striking differences are immediately apparent:

(a) For low fields, there is very little field dependence, and in particular, there is little variation *odd* in field.
netic phase transition, near 0.5 T.
(c) For fields above the phase transition, there is a gradual variation which tends to a small slope at high fields for both $\Delta l_\parallel/l_\parallel$ and $\Delta l_\perp/l_\perp$.

All of these features can be understood in the light of our model. As in the previous case, we first find the magnetostrictive strains by minimizing the magnetoelastic energy. We find

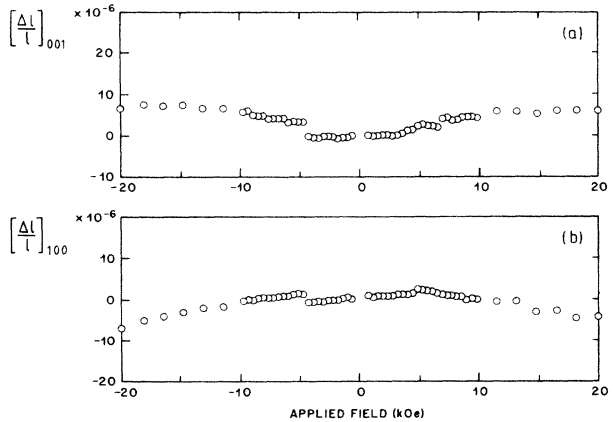

FIG. 2. Magnetostriction of DyAlG as a function of magnetic field applied parallel to [001] at $T=1.35$ K. (a) Strain measured parallel to the field $(\Delta l/l)_{[001]}$. (b) Strain measured along one of the two fourfold axes perpendicular to [001], $(\Delta l/l)_{[100]}$. After Ref. 13.

$$\epsilon_{xx}=\epsilon_{yy} = -\frac{A_1\eta^2+A_3M^2+a_3MH}{C_{11}+2C_{12}} + \frac{C_{12}(A_4M^2+a_4MH)}{(C_{11}-C_{12})(C_{11}+2C_{12})}\ , \tag{17a}$$

$$\epsilon_{zz} = -\frac{A_1\eta^2+A_3M^2+a_3MH}{C_{11}+2C_{12}} - \frac{(C_{11}+C_{12})(A_4M^2+a_4MH)}{(C_{11}-C_{12})(C_{11}+2C_{12})}\ , \tag{17b}$$

$$\epsilon_{xy} = -\frac{A_9\eta M+a_9\eta H}{C_{44}}\ , \tag{18a}$$

$$\epsilon_{yz}=\epsilon_{zx}=0\ , \tag{18b}$$

where we have omitted terms involving $\Delta\eta=\eta_x-\eta_z=\eta_y-\eta_z$, since these will only be important over a small region of field and temperature (in the antiferromagnetic phase close to the phase boundary at temperatures which are not too low[17]). In a more complete analysis we would have to include these terms.

Using Table V we see immediately that the magnetostriction $\Delta l_\parallel/l_\parallel$ is given by

$$\frac{\Delta l_\parallel}{l_\parallel} = (\epsilon_{zz})_H-(\epsilon_{zz})_{H=0} = -\frac{A_1\{[\eta(H)]^2-[\eta(0)]^2\}+A_3M^2+a_3MH}{C_{11}+2C_{12}} - \frac{(C_{11}+C_{12})(A_4M^2+a_4MH)}{(C_{11}-C_{12})(C_{11}+2C_{12})}\ . \tag{19a}$$

Similarly, for $\Delta l_\perp/l_\perp$ we find

$$\frac{\Delta l_\perp}{l_\perp} = -\frac{A_1\{[\eta(H)]^2-[\eta(0)]^2\}+A_3M^2+a_3MH}{C_{11}+2C_{12}} + \frac{C_{12}(A_4M^2+a_4MH)}{(C_{11}-C_{12})(C_{11}+2C_{12})}\ , \tag{19b}$$

which has the same functional form as $\Delta l_\parallel/l_\parallel$. Since the effects are both quite small, it is difficult to fit the parameters accurately, but we can make rough estimates.

For the asymptotic high-field behavior, we estimate from the data

$$\frac{\Delta l_\parallel}{l_\parallel}\approx(4.0+2.0\,|H|)\times10^{-6} \tag{20a}$$

and

$$\frac{\Delta l_\perp}{l_\perp}\approx(5.0-5.2\,|H|)\times10^{-6}\ , \tag{20b}$$

while at low fields there is relatively little variation.

We can thus equate

$$\frac{A_1-A_3}{C_{11}+2C_{12}}-\frac{(C_{11}+C_{12})A_4}{(C_{11}-C_{12})(C_{11}+2C_{12})}\approx 4.0\times 10^{-6}\ , \tag{21a}$$

$$\frac{A_1-A_3}{C_{11}+2C_{12}}+\frac{C_{12}A_4}{(C_{11}-C_{12})(C_{11}+2C_{12})}\approx 5.0\times 10^{-6}\ , \tag{21b}$$

$$-\frac{a_3}{C_{11}+2C_{12}}-\frac{(C_{11}+C_{12})a_4}{(C_{11}-C_{12})(C_{11}+2C_{12})}\approx 2.0\times 10^{-6}\ , \tag{21c}$$

$$-\frac{a_3}{C_{11}+2C_{12}}+\frac{C_{12}a_4}{(C_{11}-C_{12})(C_{11}+2C_{12})}\approx -5.2\times 10^{-6}\ . \tag{21d}$$

From Eqs. (21a) and (21b) we estimate

$$\frac{A_4}{C_{11}-C_{12}}\approx 1.0\times 10^{-6}\ , \tag{22a}$$

$$\frac{A_1-A_3}{C_{12}}\approx\left[\frac{C_{11}+C_{12}}{C_{12}}5.0+4.0\right]\times 10^{-6}\ . \tag{22b}$$

Similarly from Eqs. (21c) and (21d) we estimate

$$\frac{a_4}{C_{11}-C_{12}}\approx -7.2\times 10^{-6}\ , \tag{22c}$$

$$\frac{a_3}{C_{12}}\approx\left[\frac{C_{11}+C_{12}}{C_{12}}5.2-2.0\right]\times 10^{-6}\ . \tag{22d}$$

Using the measured elastic constants [Eq. (15)], we can thus find

$$A_1-A_3=2.7\times 10^6\ \mathrm{J\,m^{-3}}\ , \tag{23a}$$

$$A_4=0.2\times 10^6\ \mathrm{J\,m^{-3}}\ , \tag{23b}$$

$$a_3=2.1\times 10^6\ \mathrm{J\,m^{-3}\,T^{-1}}\ , \tag{23c}$$

$$a_4=-1.8\times 10^6\ \mathrm{J\,m^{-3}\,T^{-1}}\ , \tag{23d}$$

and, using the previously found value for $A_1=3.7\times 10^6$ $\mathrm{J\,m^{-3}}$, we can deduce

$$A_3=1.0\times 10^6\ \mathrm{J\,m^{-3}}\ . \tag{23e}$$

These values are all comparable with those for A_1, a_7, and a_9 found previously [Eq. (16)]. It should be cautioned, however, that the uncertainties in all of these parameters are really quite large and, in the absence of a more detailed analysis, they should really be regarded as no more than order of magnitude estimates.

At low fields, the variation will be more complicated, since M and η both vary, and we should also include the terms in $\eta_x-\eta_z$ and $\eta_y-\eta_z$ which we have so far omitted. However, the above expressions show that the contributions from all of the terms will in fact be quite small, since $M\ll 1$, and η changes only slowly for small fields. This agrees with the observations. The theory also shows that there should be no term linear in H for either $\Delta l_\parallel/l_\parallel$ or $\Delta l_\perp/l_\perp$. This is consistent with the experimental results in the former case and in approximate agreement in the latter case. The linear variation which is observed for $\Delta l_\perp/l_\perp$ can be explained by a small misalignment of the strain gauge.

We shall discuss all of these effects, together with additional magnetostriction data elsewhere.[13] For now, we note simply that the general theory which we have developed provides a simple model, in terms of which the field dependence at both low and high fields can be fitted.

B. Magnetoacoustic effects

1. Experimental results

When an acoustic wave is transmitted through a crystal such as DyAlG, the transit time τ is affected by three effects: magnetostrictive changes in the path length $\Delta l/l$, changes in the elastic constants $\Delta C/C$, and changes in the density $\Delta\rho/\rho$ due to magnetostriction, as discussed in Sec. II E. The last can be expressed as a corresponding change in volume $\tilde{V}=\Delta V/V$, so that

$$\frac{\Delta\tau}{\tau}=\frac{\Delta l}{l}-\frac{1}{2}\frac{\Delta C}{C}-\tfrac{1}{2}\tilde{V}\ . \tag{24}$$

All three effects will generally depend on magnetic field. It is sometimes possible to neglect the two magnetostrictive effects relative to the magnetoacoustic changes[21] but in general one must consider both contributions. In the present case, the two magnetostrictive effects turn out to be small, but not completely negligible.

Figure 3 shows the results of some recent magnetoacoustic experiments on DyAlG, taken from Ref. 8. Both the magnetic field and the direction of acoustic propagation were here parallel to [001]. A number of striking features are immediately evident.

(a) For $T=2.60$ K, which is above the Néel temperature ($T_N=2.53$ K), the changes in $\Delta\tau/\tau$ are very different for the longitudinal (L) and transverse (T) modes. For both modes, the initial variation is quadratic in field, but the high-field dependence tends toward a linear limit for the longitudinal mode and to a small, almost constant limit for the transverse mode.

(b) For temperatures below T_N the results are again quite different. First, one may note that the signs of the changes are different for the two types of modes. For the longitudinal mode, the initial curvature has the opposite sign compared to that for $T>T_N$, but the high-field limit is qualitatively similar. For the transverse mode, the initial variation is now approximately linear in field, but above a certain field there is very little variation. For both $T=1.906$ K and $T=0.888$ K, the high-field values of $\Delta\tau/\tau$ are close to those for zero field. All of the curves for $T<T_N$ show characteristic kinks, which have previously been identified with the field-induced phase transition,[8] well known in DyAlG.[9]

All of these features can be understood in terms of the magnetoelastic model developed above. We first note that the absolute magnitudes of $\Delta\tau/\tau$ are comparable with those found for magnetostriction with $\mathbf{H}\|[001]$.[13] To estimate the magnetoelastic change $\Delta C/C$ we must

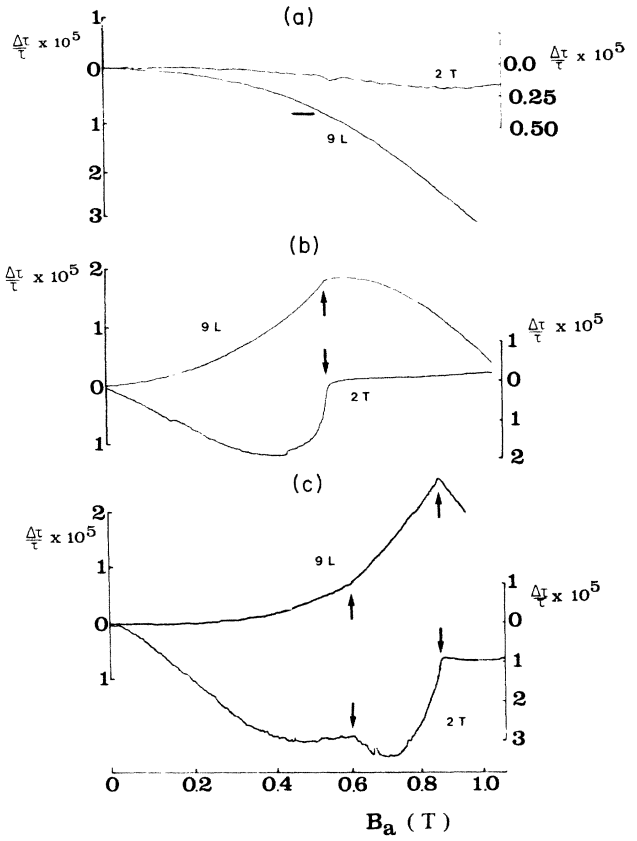

FIG. 3. Magnetic field dependence of the change of acoustic transit time, $\Delta\tau/\tau$, for the ninth longitudinal (9L) and second transverse (2T) echoes in DyAlG. (a) $T=2.60$ K $>T_N$; (b) $T=1.906$ K $<T_N$; (c) $T=0.888$ K $<T_t$. Arrows mark the critical fields corresponding to the magnetic phase boundaries. After Ref. 8.

therefore consider the possibility of a significant magnetostrictive correction to $\Delta\tau/\tau$.

2. Magnetostrictive corrections

As we have seen, there are two effects: $\Delta l/l$ and $-\frac{1}{2}\tilde{V}$. When, as in the present case, the field and the acoustic propagation directions are parallel, we can write the magnetostrictive correction terms in the form

$$\left[\frac{\Delta\tau}{\tau}\right]_{\rm MS}=\frac{\Delta l_{\parallel}}{l_{\parallel}}-\frac{1}{2}\left[\frac{\Delta l_{\parallel}}{l_{\parallel}}+2\frac{\Delta l_{\perp}}{l_{\perp}}\right]=\frac{1}{2}\frac{\Delta l_{\parallel}}{l_{\parallel}}-\frac{\Delta l_{\perp}}{l_{\perp}}\ , \tag{25}$$

and we can use the results of Fig. 2 to estimate the magnitude of the effect. For high fields, we can use the asymptotic expressions Eq. (20a) and (20b) which give

$$\frac{1}{2}\frac{\Delta l_{\parallel}}{l_{\parallel}}-\frac{\Delta l_{\perp}}{l_{\perp}}=-(3-6.2\,|H\,|)\times10^{-6}\ . \tag{25a}$$

At $H=1$ T, the correction will thus be $\sim-3.2\times10^{-6}$ and at lower fields, it will be less. It will also be smaller at higher temperatures, since both η and M will then be smaller. Compared to the measured changes in transit time, which range from $+30\times10^{-6}$ to -30×10^{-6}, the effect is thus seen to be relatively small and for the present discussion we shall neglect it. It should be noted, however, that for a detailed analysis the magnetostrictive corrections are not entirely negligible.

3. Magnetoelastic effects

If we neglect the magnetostrictive corrections, we can analyze the measured field dependence of $\Delta\tau/\tau$ in terms of the theory for $\Delta C/C$ developed in Sec. II D.

a. Longitudinal mode. For propagation along [001], we need only one second-order magnetoelastic constant, ΔC_{33}, and from Table VI, we see that

$$\frac{\Delta v}{v}=\frac{1}{2}\frac{\Delta C_{33}}{C_{11}}\ . \tag{26}$$

The dependence of ΔC_{33} on the magnetic order parameters and the field can be found from Eq. (4),

$$\Delta C_{33}=B_i\eta^2+B_3(M_x^2+M_y^2+M_z^2)+B_{12}M_z^2+b_3(M_xH_x+M_yH_y+M_zH_z)+b_{12}M_zH_z\ , \tag{27}$$

where we again omit terms in $\eta_x-\eta_z$ and $\eta_y-\eta_z$ which will be important only at intermediate fields. For $\mathbf{H}\|[001]$, we put $M_x=M_y=0$, $M_z=M$, and Eq. (27) can then be written in the form

$$\frac{1}{2}\frac{\Delta C_{33}}{C_{11}}=a_{\rm L}M^2+b_{\rm L}MH+c_{\rm L}\eta^2\ . \tag{28}$$

At high fields, this tends to the asymptotic form $a_{\rm L}+b_{\rm L}H$, while at low fields it tends to $c_{\rm L}\eta^2$. The change between high and low fields is thus

$$\frac{1}{2}\left[\frac{\Delta C_{33}}{C_{11}}\right]_{H\to\infty}-\frac{1}{2}\left[\frac{\Delta C_{33}}{C_{11}}\right]_{H=0}=(a_{\rm L}-c_{\rm L}\eta^2)+b_{\rm L}H\ . \tag{28a}$$

If we examine the results in Fig. 3, we see that the high-field variation of $\Delta\tau/\tau$ does indeed appear to be linear, and for the three temperatures $T=2.6$, 1.9, and 0.89 K, we can estimate

$$-(a_{\rm L}-c_{\rm L}\eta^2)\times10^5=3.3,\ 6.5,\ \text{and}\ 9.9\ , \tag{29a}$$

$$b_{\rm L}\times10^5=6.5,\ 6.0,\ \text{and}\ 8.3\ , \tag{29b}$$

respectively. We see that $b_{\rm L}$ is roughly independent of temperature, as the theory predicts, but the zero-field intercept varies markedly with temperature. For $T=2.6$ K, $\eta=0$, so that we can identify $a_L=-3.3\times10^{-5}$, while at $T=0.89$ K, $\eta\approx1$, which gives $c_L=6.6\times10^{-5}$. The intermediate value for $T=1.90$ K then corresponds to $\eta\approx0.7$, a value which is not unreasonable, since $T_N=2.53$ K.

We can use this analysis to predict the upward curva-

ture observed at $T=1.90$ K. If we consider $\Delta\tau/\tau$ at the phase boundary, we see that we would expect a change from the zero-field value of

$$\left[\frac{\Delta\tau}{\tau}\right]_{H=H_C}=-(a_L M_C^2-c_L\eta^2+b_L H_C M_C)\ , \tag{30}$$

where M_C and H_C are the values at the second-order phase boundary. From magnetization data,[11] we know that at $T=1.90$ K, $M_C\approx 0.52$, and $H_C\approx 0.55$ T. Thus, we would expect

$$\left[\frac{\Delta\tau}{\tau}\right]_{H=H_C}\approx[3.3\times(0.52)^2+6.6\times(0.7)^2 -6.0\times 0.55\times 0.52]\times 10^{-5}$$

$$=2.4\times 10^{-5}\ , \tag{30a}$$

in excellent agreement with the experimental value of 1.8×10^{-5}.

We thus see that the variation of $\Delta\tau/\tau$ for the longitudinal mode can be explained in terms of three contributions: positive contributions proportional to M^2 and η^2 and a negative contribution in MH. The observed variation is the result of a delicate competition between them. It should again be emphasized that the quantitative aspects of the analysis which we have given have large uncertainties, which are difficult to estimate, and a much more detailed discussion, with results from many more temperatures and more quantitative estimates of M and η as functions of T and H, is really needed to determine the parameters. The small magnetostriction corrections should then also be included.

b. Transverse modes. There are two transverse modes which, as we have seen in Sec. II E, will not be degenerate in the present geometry. Since the plane of polarization of the transducer relative to the principal axes [110] and [1$\bar{1}$0], was not specified in Ref. 8, we cannot tell exactly what polarization was actually measured. If the angle of the generator or detector relative to [110] is θ, the signal will be proportional to

$$\frac{\Delta v}{v}=\cos^2\theta\frac{\Delta v_1}{v_1}+\sin^2\theta\frac{\Delta v_2}{v_2}\ , \tag{31}$$

which can vary between $\Delta v_1/v_1$ and $\Delta v_2/v_2$. In terms of the magnetoelastic parameters,

$$\frac{\Delta v_1}{v_1}=-\frac{1}{2C_{44}}(\Delta C_{44}+\Delta C_{45})\ , \tag{32a}$$

$$\frac{\Delta v_2}{v_2}=-\frac{1}{2C_{44}}(\Delta C_{44}-\Delta C_{45})\ , \tag{32b}$$

so that

$$\frac{\Delta v}{v}=\frac{1}{2C_{44}}(\Delta C_{44}+\alpha\Delta C_{45})\ , \tag{33}$$

where $\alpha=\cos^2\theta-\sin^2\theta$, which can vary between ± 1. From Tables II and IV, we see that

$$\frac{1}{2}\frac{\Delta C_{44}}{C_{44}}=a_T M^2+b_T M_z H_z+c_T\eta^2\ , \tag{34a}$$

similar to the form for ΔC_{33}, but that

$$\frac{1}{2}\frac{\Delta C_{45}}{C_{44}}=d_T M_z\eta+e_T H_z\eta \tag{34b}$$

has a very different variation, depending *linearly* on η. Here we have again omitted the terms in $\eta_x-\eta_z$ and $\eta_y-\eta_z$.

From Eqs. (33) and (34b), we see immediately that $\Delta v/v$ will generally vary linearly with field at low fields when $\eta\neq 0$, i.e., below T_N, but quadratically when $T>T_N$, as observed experimentally. The slope at lowest temperatures and low fields can be interpreted in terms of the single term αe_T, since M is small and $\eta\rightarrow\pm 1$ as $T\rightarrow 0$ K. From Fig. 2, we find $\Delta\tau/\tau\sim -5.6H\times 10^{-5}\approx\pm\alpha e_T H$. At higher temperatures, we must also expect a contribution from the term in αd_T and, estimating $\partial M/\partial H$ at $T=1.9$ K from magnetization data[11] and $|\eta|=0.7$ from the magnetostriction analysis, we find from the slope at $T=1.906$ K:

$$\alpha(d_T M+e_T H)\eta=\alpha(0.44d_T+e_T)\times 0.7H\ ,$$

which can be equated to the experimental value, $\mp(3.1\times 10^{-5})H$. Hence, $\alpha d_T=\pm 2.5\times 10^{-5}$, and $\alpha e_T\mp 5.6\times 10^{-5}$. To relate d_T and e_T to the corresponding terms B_{28} and b_{28} in Tables II and IV, we would have to know the polarization factor and on the basis of the present experiments, all we can conclude is that

$$|B_{28}|=|2C_{44}d_T|\geq 5.6\times 10^6 \tag{35a}$$

$$|b_{28}|=|2C_{44}e_T|\geq 12.4\times 10^6\ , \tag{35b}$$

with the equality corresponding to the possibility $\alpha=1$ for the experiment cited. Further experiments are needed to resolve this.

The ambiguity in sign corresponds to the uncertainty concerning the sign of η. The absolute sign is difficult to determine but its relative sign can be manipulated by means of a suitable field cycle.[25] Our theory predicts, therefore, that $\Delta\tau/\tau$ should change sign when the sign of η is reversed. Such an effect has indeed been observed and will be reported elsewhere.[10] The theory also predicts, conversely, that there should be *no* change of sign for the longitudinal mode, and this too has been confirmed experimentally.[10]

For high magnetic fields or for $T>T_N$, $\eta=0$ and Eqs. (33) and (34) predict that $\Delta\tau/\tau$ should vary as

$$-(a_T-c_T\eta^2)_{H=0}-b_T H_z\ .$$

The experimental results are not inconsistent with this, but the coefficients a_T, b_T, and c_T are evidently all very small ($<\sim 1\times 10^{-5}$). It remains as a challenge for the microscopic theory to show why a_T, b_T, and c_T should be small, while a_L, b_L, and c_L were large.

The only other experimental feature which is not readily explained by the theory is the dip in the

transverse-mode field sweep, which is observed in the mixed phase region at the lowest temperature, indicated by the arrows near 0.6 and 0.85 T. For the longitudinal mode, the behavior is as one might expect, with a linear interpolation between the two and end points, corresponding to the change in the proportion of the antiferromagnetic and paramagnetic phases as the field is increased. For the transverse mode, the behavior is clearly more complicated. One can speculate that this may somehow be related to the fact that there are two transverse modes whose principal axes depend on the sign of η and on magnetostrictive strain. It seems possible that this situation might be complicated in the mixed phase state, in which there are microscopic domains. Further studies are clearly needed to resolve this.

In general, however, we now have a good understanding of the overall features of the magnetoacoustic effects which have been observed. Our theory makes many additional predictions for other field orientations and for other directions of propagation of the acoustic waves and it will be interesting to compare these with future experiments.

C. Piezomagnetic effects

1. Strain-induced magnetization

The induction of a magnetic moment through the application of an elastic strain has been studied extensively in various tetragonal and rhombohedral crystals,[26] but it has previously not been observed in cubic antiferromagnets. For the effect to occur one needs a term in the energy of the form

$$E_{\mathrm{ME}}=K_{pq\mu}\epsilon_{pq}M_{\mu}\ , \tag{36}$$

where $p,q,\mu=x,y,z$ with $K_{pq\mu}\neq 0$. If K is simply a constant, such a term is clearly forbidden by time-reversal symmetry. However, if K contains as a factor an antiferromagnetic order parameter which is itself odd under time reversal, such a term may be allowed.[27,28] In many materials, however, the antiferromagnetic order involves an enlargement of the unit cell, so that a term of the form of Eq. (36) is still not allowed because M_{μ} and ϵ_{pq} are translationally invariant and K is not. Only in antiferromagnets in which the order can be described within the crystallographic unit cell can a term of the form of Eq. (36) be an invariant. DyAlG is one of the materials where this is the case.

From Table I, we see that there are indeed terms in the energy of the form

$$E_{\mathrm{ME}}=A_9\eta(\epsilon_{xy}M_z+\epsilon_{yz}M_x+\epsilon_{zx}M_y) \tag{37}$$

which will give rise to some unusual effects.

We have already noted that the application of a field along [001] produces an orthorhombic magnetostrictive distortion with $\epsilon_{xy}\neq 0$. Such a distortion would not be expected in a normal cubic crystal for which the [001] axis has fourfold symmetry.

The terms in Eq. (37) can similarly lead to the inverse effect: The application of a nonzero strain in the antiferromagnetic state ($\eta\neq 0$) will induce a magnetization, just as a magnetic field. The strain could be externally applied, or it could be an internal strain produced in the preparation of the sample.

It is not easy to calculate the size of the effect, but we can make a very rough estimate of the order of magnitude we might expect. If we make the reasonable assumption that all of the terms in the energy expansion which are not identically zero have roughly the same order of magnitude, as was shown to be the case where we were able to estimate magnitudes [Eq. (23)], we can immediately conclude that the induced moment should be roughly comparable to the corresponding strain. Strains due to crystal imperfections are generally quite small, so that we would not expect any large moments in zero field.

Nevertheless, this effect does provide a mechanism for a nonzero moment and the corresponding magnetic hysteresis. This may help to explain the anomalous effects which have been reported for small particles of DyAlG which show just such a small ($M\sim 10^{-4}$) permanent moment.[11,12]

2. Strain-induced antiferromagnetic order

An even more striking consequence of the piezomangetic coupling may be expected near the second-order transition in the presence of a magnetic field. Here nonzero strains combine with the components of the magnetization to provide an induced staggered field, H_s, which acts on the antiferromagnetic order parameter

$$(A_9\epsilon_{pq}M_{\mu})\eta=-H_s\eta\ . \tag{38}$$

Such an induced staggered field was previously recognized by Blume *et al.*[29,30] to arise from a higher-order coupling term of the form $M_xM_yM_z\eta$, and the effect of such a term has been studied extensively.[31] However, whenever any one of the components of M is zero, this effect vanishes and there should then be no staggered field. It has been puzzling for some time that there are, nevertheless, induced staggered field effects even in cases where one or two of the M's are zero, in particular for $\mathbf{H}\|[001]$ and $\mathbf{H}\|[110]$.[29,32] There have been attempts to explain the observations in terms of the coupling of other order parameters and the possibility of nonlocalized spin densities.[16] However, none of these explanations could be confirmed quantitatively and there is certainly room for an alternative mechanism.

One might assume that the piezomagnetic effect would once again be very small for reasonable strains, as it was in the zero-field case. However, there is an important difference, inasmuch as the staggered susceptibility $\partial\eta/\partial H_s$ *diverges* as the second-order transition is approached. Even a small staggered field will, therefore, produce a significant effect.

If we consider the effect *at* the second-order phase boundary, and use Landau theory to estimate η as a function of H_s, we note that η is proportional to $H_s^{1/3}$, as in the usual case of a critical isotherm. The presence of the cube root will clearly enhance the effect of even a small staggered field, and it is not unreasonable to conclude that even small strains could account for the small

amount of antiferromagnetic order observed just above the nominal phase boundary. (In a more accurate theory the factor $\frac{1}{3}$ would be replaced by $1/\delta$, where $\delta \sim 5$, so that the effect would be even more pronounced.)

The piezomagnetic coupling may also be expected to have a significant effect near a tricritical point, where both the staggered and the uniform susceptibilities will diverge. It could well be that the extreme experimental difficulties which have been experienced in high-resolution studies of the tricritical point of DyAlG (Ref. 33) have been due, in part, to magnetoelastic effects and small random strains. More quantitative studies are needed to explore these ideas further.

IV. MICROSCOPIC MECHANISMS

We have left until now the consideration of microscopic mechanisms, because this is clearly quite a complicated problem and it is helpful to have the guidance of experimental results.

There are at least five mechanisms which may be important. Four of the mechanisms involve the strain modulation of the magnetic dipole and exchange couplings, each through two mechanisms: variation of the separations between the spins, and changes in the ionic wave functions. The latter can be thought of partily as a change in the g values, and this gives rise to an additional mechanism: a change in the direct interaction with a magnetic field. It is clear that quantitative estimates will be rather uncertain, since the five mechanisms can compete, with positive and negative contributions. There are, however, a number of features which can be recognized.

A. Modulation of dipole and exchange interactions

One effect which can be calculated exactly is the strain modulation of the magnetic dipole coupling through the variation of lattice distances. We have calculated the corresponding contributions to the A_n but we shall not give the details here, since the effects all turned out to be small. The results may be found in Ref. 34.

A more important contribution may come from the corresponding modulation of the exchange interaction. For nearest neighbors this is known to be about one half of the magnetic dipole coupling,[35] but its dependence on separation will be considerably more rapid than the r^{-3} dependence for magnetic dipoles.

One can obtain a rough estimate of the sum of the two effects by a simple order of magnitude calculation. The magnetic energy difference between the ordered ($\eta=1$) and disordered ($\eta=0$) states in zero field has been found, using heat capacity measurements,[36] to be

$$U_M = -1.92R = -3.6\times 10^5 \text{ J m}^{-3} , \tag{39}$$

where $R=8.31$ J/mol, and where we use the molar volume $V_M=43.8$ cm^3.[36] If the lattice is now strained, for example by a uniform compression, the change in energy will clearly be of order

$$\delta E = pu_M(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})\eta^2 , \tag{40}$$

where p is a factor which would be -3 for pure dipole coupling and maybe -20 for exchange coupling. Thus, taking $p\sim -10$ we might expect the parameter $A_1 = pU_M$ to be of order 3×10^6 J cm^{-3}. The experimental estimate is, in fact, 3.7×10^6 J m^3, in surprisingly close agreement.

Similar considerations suggest that the other parameters A_n should all be of roughly the same order of magnitude, since they all represent different aspects of the same basic effect, the strain modulation of the magnetic energy. Experimentally, this seems to be the case.

We can also make rough estimates of the B_n in the same way. Since the B_n's involve second derivatives with respect to lattice distance, we would expect the factor p in (40) to be replaced by $p(p-1)/2$. Thus, $B_n/A_n \sim (p-1)/2$ and for $p=-10$, this gives a factor of -5.5. This is consistent with our estimates for a_L, b_L, and c_L which correspond to magnetoelastic parameters ranging from -33 to 66×10^6, an order of magnitude larger than the A_n.

One puzzle to be resolved is the smallness of the magnetoelastic parameters corresponding to a_T, b_T, and c_T. These all involve second-order shear strains, and there is no obvious reason why they should be much smaller than the terms involving compressional strains. A more detailed calculation would be of interest, but it will not be easy to parametrize the strain dependence of the exchange interactions, which are quite complicated in this system.

B. Strain dependence of Zeeman interactions

The existence of terms linear in the field, linear in the strain, and linear in one of the order parameters can not be explained in terms of a modulation of the spin-spin interactions. For this we must invoke a change in the g values resulting from a change in the crystal field. The detailed calculation of such an effect will be quite complicated, but we can again derive a simple order of magnitude estimate.

In general, a strain ϵ_{pq} will result in a change of the crystal field of the order of $\delta V_c = \epsilon_{pq} V_c^0$ is the unperturbed crystal field. δV_c will contain a number of different terms, as allowed by the symmetry, and each of these will result in an admixture into the ground state of order $\delta V_c/V_c^0$ i.e., $\sim\epsilon_{pq}$. Consequently, the g tensor of the ground state will change by $\delta \mathrm{g} \sim \epsilon_{pq} \mathrm{g}^0$, where g^0 is the unperturbed g tensor. For DyAlG g is known to be Ising-like with one large component parallel to a local z axis.[35] The *change* δg, however, does not have to be Ising-like and in particular, it can have off-diagonal components relative to the unperturbed principal axes.

In the presence of a large field with components H_x, H_y, and H_z relative to the *local* axes, the energy of the ground state in the unperturbed state is given by

$$E^0 = -\tfrac{1}{2} g_z \mu_B H_z . \tag{41}$$

When a strain is applied, the energy will change to

$$E^0+\delta E=-\frac{\mu_B}{2}\{[\delta g_{xz}H_x+\delta g_{yz}H_y+(g_{zz}+\delta g_{zz})H_z]^2 +(H_x\delta g_{xx}+H_y\delta g_{yx}+H_z\delta g_{zx})^2 +(H_x\delta g_{xy}+H_y\delta g_{yy}+H_z\delta g_{zy})^2\}^{1/2} . \quad (42)$$

If the $\delta g_{\alpha\beta}$ are very small, we can expand Eq. (42) to obtain the leading terms

$$\delta E=-\frac{\mu_B}{2}(\delta g_{xz}H_x+\delta g_{yz}H_y+\delta g_{zz}H_z) , \quad (43)$$

which, since $\delta g_{\alpha\beta}\sim\epsilon_{pq}$, provide the required coupling mechanism between the strain and the field. To find the corresponding energy density, we must sum over the different sites in the unit cell and we must also allow for thermal excitation when $M\neq 1$. The resulting magnetoelastic energy has the form

$$E_{\rm ME}=\sum_{p,q,\mu,j}F_{pq\mu j}\epsilon_{pq}H_\mu G_j , \quad (44)$$

where $p,q,\mu=x,y$, or z and G_j is one of the six order parameters Eq. (1). This expression has the form of the terms in Table III and the parameter $F_{pq\mu j}$ will be governed by the same overall symmetry considerations.

The advantage of the microscopic derivation is, of course, that we can now estimate the order of magnitude of F's. Using a typical value of 5×10^5 J T^{-1} m^{-3} for the magnetization (500 emu/cm^3), we see that the F's will have values which are comparable with the a_n deduced from the magnetostriction data. Of course, there are many numerical factors of order unity which we have omitted and which can change the final result by a factor of 10 or more, but the order of magnitude of the present estimate is quite satisfactory.

It is easy to see physically how a magnetic field will couple to the shear strains in a system such as DyAlG. Whenever the field is not parallel to the local z axis of a particular Dy^{3+} ion, there will be a torque which will distort the environment of the ion and lead to a microscopic shear strain. Since the local axes point in different directions, the strain in the presence of a field will never be zero. This effect is thus the result of the large local anisotropy, combined with the noncollinear structure of DyAlG.

The coupling to the compressional strains is similar, but somewhat more subtle. Components parallel to one of the local axes will simply produce changes in the crystal field but no change in the point symmetry. Thus, no off-diagonal terms in δg are produced. However, since two of the three local axes at each site are not parallel to the cubic crystal axes,[37] even a uniform compressional strain can not coincide with the local axes at all sites, and a reduction in point symmetry at some sites will always result.

Thus all components of strain will lead to off-diagonal components in δg and a coupling to field of the form we have postulated.

The higher-order terms in the expansion of Eq. (42) also provide the magnetoelastic energy quadratic in the strains and linear in the field (Table IV). The order of magnitude of the coefficients will again be comparable to the observed values.

The strain-induced changes in the g values will also, of course, contribute to the strain modulation of the spin-spin interactions. The orders of magnitude will be similar to those arising from the variation of the lattice distances considered in the previous section. There is no *a priori* way of predicting whether the effects will add or cancel. Clearly, it would be of interest to carry out a detailed calculation of all these effects, using the known wave functions and energy levels,[38] but this is a formidable task which we have not yet attempted.

V. CONCLUSION

We have seen that magnetoelastic effects can lead to some unusual and interesting phenomena in DyAlG. The analysis is complicated by the large number of terms which are allowed by the garnet structure, but we have been able to reach a number of quantitative, as well as some qualitative conclusions. In particular, we have been able to interpret some striking features observed in the field and temperature dependence of both magnetostriction and magnetoacoustic experiments.

The most interesting effects arise from a property of the structure which will also occur in many other antiferromagnets: the fact that the order parameter is translationally invariant and not, as is more often the case, associated with a finite wave vector. This simple fact alone gives rise to the possibility of a variety of terms in the energy coupling the antiferromagnetic order to elastic strains, the magnetic field, the magnetization, and to other order parameters, with a resulting richness in behavior. Such coupling terms are clearly possible also in other simpler antiferromagnets, provided only that the magnetic and chemical unit cells coincide. Under these conditions, we must expect magnetoelastic effects to affect the usual magnetic properties in nontrivial ways.

The most unexpected effect which we have found is the coupling between the antiferromagnetic order and shear strains which will act as an induced staggered field in the presence of a uniform magnetic field. Such an effect will, of course, destroy the actual phase transition to the paramagnetic state, since there will always be some small amount of antiferromagnetic order for all strengths of magnetic field.

For other magnetic systems, our findings emphasize the possible importance of elastic effects in general and the possibility of significant consequences from relatively small elastic strains. It would seem clear that such effects must be considered very carefully in all high-resolution studies, and especially near critical and multicritical points.

In any case, we must conclude that elastic effects can not be neglected in the detailed understanding of model magnetic materials.

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