

Spin waves in terbium. II. Magnon-phonon interaction

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The selection rules for the linear couplings between magnons and phonons propagating in the c direction of a simple basal-plane hcp ferromagnet are determined by general symmetry considerations. The acoustic-optical magnon-phonon interactions observed in the heavy-rare-earth metals have been explained by Liu as originating from the mixing of the spin states of the conduction electrons due to the spin-orbit coupling. We find that this coupling mechanism introduces interactions which violate the selection rules for a simple ferromagnet. The interactions between the magnons and phonons propagating in the c direction of Tb have been studied experimentally by means of inelastic neutron scattering. The magnons are coupled to both the acoustic- and optical-transverse phonons. By studying the behavior of the acoustic-optical coupling, we conclude that it is a spin-mixed-induced coupling as proposed by Liu. The coupled magnon-transverse-phonon system for the c direction of Tb is analyzed in detail, and the strengths of the couplings are deduced as a function of wave vector by combining the experimental studies with the theory.

I. INTRODUCTION

The large coupling between the lattice and the spin system which has been observed in many of the heavy-rare-earth metals is due to the large values of the orbital momentum \vec{L} of the ions (with the exception of Gd). The spin-lattice coupling may be considered as a special type of magnetic anisotropy, and the present paper is closely connected with the preceding and the subsequent papers, to be referred to as I and III, where other aspects of magnetic anisotropy in Tb are considered (I: two-ion magnetic anisotropy, III: magnetic anisotropy at zero wave vector).

If the magnetic moments are ordered, the coupling between the lattice and the spin system may manifest itself as a distortion of the lattice, and as couplings between the lattice vibrations and the magnetic excitations. We begin connecting the static and dynamic phenomena by a generalization of the static Hamiltonian¹ to the dynamic case. Through this approach, we may utilize the detailed knowledge of the static Hamiltonian (III) in a discussion of the dynamic behavior of the coupled systems. The frozen-lattice model for the energy gap in the spin-wave spectrum at zero wave vector, which is considered in detail in III, is found to be correct within the harmonic approximation. In continuation of the discussion in I, we argue that higher-order magnetoelastic contributions to the \vec{q} -dependent magnetic anisotropy in Tb are of minor importance.

From a general spin-lattice Hamiltonian linear in the ionic displacements, we deduce the selection rules for the direct couplings between magnons and phonons propagating in the c direction of a basal-plane ferromagnet. The magnons interact

both with the acoustic and optical transverse phonons. The acoustic-optical coupling has been explained by Liu² as being due to the spin-orbit coupling of the conduction electrons. The selection rules deduced for the interactions arising from the mechanism proposed by Liu are found to differ from those valid for the coupling in a simple ferromagnet. By studying the transverse phonon spectrum in the c direction of Tb by inelastic neutron scattering we found that the acoustic-optical magnon-phonon interaction is entirely dominated by the spin-mixed-induced coupling, in accordance with the proposal of Liu.

The Hamiltonian for the coupled magnon-transverse-phonon system of a basal-plane ferromagnet is derived. The equations of motion for the six (different) modes propagating in the c direction are deduced. Because of the coupling, the normal modes are no longer pure magnon or phonon states, and energy gaps occur at the crossing points of the unperturbed magnon and phonon dispersion relations. From the magnitudes of these energy gaps, which were measured by inelastic neutron scattering, and from the expected behavior of these couplings we deduce the \vec{q} -dependent strength of the magnon-phonon interactions in the c direction of Tb.

II. MAGNETOELASTIC COUPLING

In III we establish the general single-ion magneto-elastic Hamiltonian linear in the strains, Eq. (6), which transforms in accordance with the point symmetry of Γ in the Brillouin zone of the hexagonal lattice.¹ If the magnetic moments are ordered, a coupling between the lattice and the spin system may be manifested through deforma-

tions of the lattice which minimize the total magneto-elastic energy. From the degree to which the lattice is deformed, the strength of the coupling may be deduced if the corresponding change in the elastic energy (the elastic constants) are known [Eqs. (8)–(10) of III]. Without a detailed consideration of their origin, which may be two as well as single ion, mediated by the Coulomb or exchange interaction, the phenomenological coupling parameters obtained in this way may serve as an adequate basis for a treatment of the dynamics of the coupled systems. Neglecting two-ion anisotropy, the magnetoelastic Hamiltonian was generalized to take into account the interactions between elastic waves and the spin waves in a basal-plane ferromagnet by Jensen,^{3,4} and this treatment was later extended to the cases of conical and helical ordering by Nayyar and Sherrington.⁵ Here we shall restrict ourselves to the case of a ferromagnet which has only one atom per unit cell. The tensor spin operators are expanded in magnon operators as considered in I [Eqs. (16)–(17)] and the deviations from the homogeneous strains may be written in terms of normal phonon coordinates by using the local strain theory of Evenson and Liu.⁶ Proceeding in this way we may write the total Hamiltonian for the spin-lattice system³

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{m-p}^{(1)} + \mathcal{H}_{m-p}^{(2)}. \quad (1)$$

The unperturbed Hamiltonian

$$\mathcal{H}_0 = E_0 + \sum_{\vec{q}} \epsilon(\vec{q}) \alpha_{\vec{q}}^\dagger \alpha_{\vec{q}} + \sum_{\vec{k}, s} \omega_s(\vec{k}) \beta_{s, \vec{k}}^\dagger \beta_{s, \vec{k}} \quad (2)$$

includes the terms arising from the homogeneous part of the strains which contribute to the energies of the magnons, $\epsilon(\vec{q})$, with the “frozen-lattice” contributions⁷ which are deduced in III. \mathcal{H}_0 is diagonal in the magnon operators $\alpha_{\vec{q}}^\dagger$ and in the phonon operators $\beta_{s, \vec{k}}$, where s denotes one of the three phonon branches. The interaction term

$$\mathcal{H}_{m-p}^{(1)} = \sum_{s, \vec{k}} i W_s(\vec{k}) (\alpha_{\vec{k}}^\dagger + \alpha_{-\vec{k}}) (\beta_{s, \vec{k}} + \beta_{s, -\vec{k}}) \quad (3)$$

involves one-magnon–one-phonon scattering processes, whereas the other interaction term represents interactions between two magnons and one phonon,

$$\begin{aligned} \mathcal{H}_{m-p}^{(2)} = & \sum_{s, \vec{k}, \vec{q}} [U_s(\vec{k}, \vec{q}) \alpha_{\vec{q}+\vec{k}}^\dagger \alpha_{\vec{q}} \\ & + V_s(\vec{k}, \vec{q}) \frac{1}{2} (\alpha_{\vec{q}+\vec{k}}^\dagger \alpha_{-\vec{q}}^\dagger + \alpha_{\vec{q}}^\dagger \alpha_{-\vec{q}-\vec{k}})] \\ & \times (\beta_{s, \vec{k}} + \beta_{s, -\vec{k}}). \end{aligned} \quad (4)$$

The derivation of the interaction amplitudes⁴ is

tedious but straightforward, and for the present purpose we shall only quote the amplitude of the direct coupling between the magnons and the transverse phonons propagating in the c direction of a basal-plane ferromagnet. The assumption of one atom per unit cell then corresponds to the use of the double-zone representation in the case of an hcp lattice. When this coupling is introduced, the double degeneracy of the transverse phonons is lifted because only the phonons with the polarization vector parallel to the magnetization may interact, and the strength of the coupling is given by

$$W(k) = \frac{1}{4} c_\epsilon H_0 \frac{\hbar}{c} \left(\frac{J\sigma}{M} \right)^{1/2} \left(\frac{\epsilon(k)}{\omega(k)[A(k)+B(k)]} \right)^{1/2} \sin \frac{1}{2} kc, \quad (5)$$

where c is the lattice parameter and M the mass of the ions. $c_\epsilon H_0$ is the magnetoelastic coupling parameter connected with the ϵ -strain deformations of the lattice as considered in III [Eqs. (5) and (24)], which depends implicitly on the relative magnetization σ . $A(k)+B(k)$ is the magnon-energy parameter defined in I [Eq. (21)]. Two-ion contributions to the coupling are effectively included to first order in kc and may only introduce deviations of the order of $(kc)^3$, from the relation $W(-\vec{k}) = -W(\vec{k})$, which justifies use of an effective single-ion Hamiltonian for the magnetoelastic couplings in at least the region of the Brillouin zone near Γ .

If $\mathcal{H}_{m-p}^{(2)}$ is neglected, the Hamiltonian (1) can be diagonalized exactly by means of a canonical transformation of the magnon and phonon operators. The normal modes are then no longer pure magnon or phonon states, and energy gaps^{3,4}

$$\Delta_s(\vec{k}) \cong 2 |W_s(\vec{k})| \quad (6)$$

may occur at the crossing points of the unperturbed magnon and phonon dispersion relations. In the long-wavelength limit, the velocity of the sound waves is changed according to

$$\omega'_s(\vec{k})/\omega_s(\vec{k}) = [1 - 4W_s^2(\vec{k})/\omega_s(\vec{k}) \epsilon(0)]^{1/2}, \quad \vec{k} \rightarrow \vec{0}, \quad (7)$$

which is independent of \vec{k} in this limit. The magnon energy gap $\epsilon(0)$ at zero wave vector is always finite if magnetoelastic couplings are present because of the frozen-lattice contributions. $\epsilon(0)$ finite and the condition $W_s^2(\vec{k}) \propto |\vec{k}|$ at small wave vectors imply that the one-magnon–one-phonon interaction $\mathcal{H}_{m-p}^{(1)}$ does not contribute to the magnon energy gap at $k=0$. This result has been obtained by several authors (Jensen,⁴ Chow and Keffer,⁸ and Liu⁹), and it supports the validity of the frozen-lattice model for the energy gap $\epsilon(0)$,

in which model the presence of phonons is neglected. Apart from the possible contributions from $\mathcal{H}_{m-p}^{(2)}$ to $\epsilon(0)$ the frozen-lattice model proposed by Turov and Sharov⁷ is found to be correct. The two-magnon-one phonon interactions may be taken into account by second-order perturbation theory and may, for instance, contribute (at finite temperatures) to the energy of the long-wavelength phonons.^{3,4} Here we are mostly concerned with the magnons, and, by summing over all virtual phonon states in (4), we transform the phonon operators into two magnon operators, which implies that the interactions in $\mathcal{H}_{m-p}^{(2)}$ are equivalent to magnon-magnon interactions and as such contribute to the energy renormalization and the lifetime of the magnons. At zero temperature $\mathcal{H}_{m-p}^{(2)}$ will introduce a zero-point correction of the magnon energies, analogous with magnon-magnon interactions,¹⁰ which is deduced⁴ to be

$$\Delta\epsilon(\vec{q}) = - \sum_{s, \vec{k}} \left(\frac{|U_s(\vec{k}, \vec{q})|^2}{\omega_s(\vec{k}) + \epsilon(\vec{q} + \vec{k}) - \epsilon(\vec{q})} + \frac{|V_s(\vec{k}, \vec{q})|^2}{\omega_s(\vec{k}) + \epsilon(\vec{q} + \vec{k}) + \epsilon(\vec{q})} \right). \quad (8)$$

However, this change of the magnon energies should be accompanied by a similar zero-point adjustment of the homogeneous strains which are present in the unperturbed magnon energies (the frozen-lattice contributions). The use of the phenomenological coupling parameters, which are determined by minimizing the free energy [Eqs. (8)-(10) of III], turns the question of a possible deviation of $\epsilon(0)$ from the frozen-lattice energy gap into the question of a possible existence of differences between static and dynamic energy-gap parameters, due to magnon-magnon interactions. These features of the dynamics of the coupled spin-lattice system may be elucidated by the following formal treatment, which also shows that phonon-induced multipole interactions [mechanism (viii) in I] are identical to magnon-phonon interactions. A simplified version of the magnetoelastic Hamiltonian may be written

$$\mathcal{H}_{m-e} = \sum_i \left(\frac{1}{2} c' e_i^2 - e_i \sum_{lm} B_{lm} \tilde{O}_{l,m}(J_i) \right), \quad (9)$$

where we consider only one strain variable e_i , which depends on the position of the i th ion. c' is a reduced elastic constant, and the Racah operators $\tilde{O}_{l,m}$, are introduced in I. Without introducing explicitly the magnon operators, this expression may be treated as above. The free energy is minimized, and the inhomogeneous part of the strain is transformed to the spin space by summing over all virtual phonon states appearing in a second-order

perturbation calculation. Equation (9) then takes the form

$$\begin{aligned} \mathcal{H}_{m-e} = & \sum_i \left(\frac{1}{2} c' \bar{e}^2 - \bar{e} \sum_{lm} B_{lm} \tilde{O}_{l,m}(J_i) \right) \\ & + \sum_{i,j} \sum_{lm} \sum_{l'm'} K_{ll'}^{mm'}(\vec{R}_i - \vec{R}_j) \\ & \times [\tilde{O}_{l,m}(J_i) - \langle \tilde{O}_{l,m}(J_i) \rangle] \\ & \times [\tilde{O}_{l',m'}(J_j) - \langle \tilde{O}_{l',m'}(J_j) \rangle], \quad (10) \end{aligned}$$

where we have assumed ferromagnetic ordering [$\langle \tilde{O}_{l,m}(J_i) \rangle$ is constant]. The first part of (10) is the frozen-lattice contribution, whereas the second part describes the dynamic interaction between the lattice and the spin system corresponding to $\mathcal{H}_{m-p}^{(1)} + \mathcal{H}_{m-p}^{(2)}$ in (1), which implies a coupling between the moments on different sites mediated through the phonons. If we neglect higher-order terms than those linear or quadratic in magnon operators in the expansion of the Racah operators [Eqs. (16) and (17) of I], it follows immediately that the quadratic terms in the second part of (10), corresponding to $\mathcal{H}_{m-p}^{(2)}$, do not contribute to the magnon energies, whereas the linear terms corresponding to $\mathcal{H}_{m-p}^{(1)}$ may give rise to dispersion. However, the linear terms do not contribute to the energy at zero wave vector because the Fourier transform of $K_{ll'}^{mm'}(\vec{R}_i - \vec{R}_j)$ vanishes at $q=0$, as the sum of the displacements of the ions from their equilibrium positions, $e_i - \bar{e}$, is zero. The last formulation of the dynamic problem, (10), shows in a transparent way that the frozen-lattice model for the magnon energy gap is correct, when higher-order effects such as magnon-magnon interactions are neglected. The correction obtained above, Eq. (8), is more fictitious than real and is compensated at $q=0$ by a corresponding change of the ground state, which is included in \bar{e} in (10). This conclusion is supported by the comparison in III between the static and dynamic energy-gap parameters obtained experimentally in Tb, where the best agreement is found for the parameter $P_0(-)$, which is entirely dominated by magnetoelastic contributions. In (10) the terms corresponding to $\mathcal{H}_{m-p}^{(2)}$ disappear if magnon-magnon interactions are neglected, whereas (8) represents a \vec{q} dependence of the frozen-lattice contributions, as $\Delta\epsilon(\vec{q})$ depends on \vec{q} . These contributions to the \vec{q} -dependent anisotropy in Tb (I) are presumably unimportant and cannot be distinguished from the equivalent contributions arising from the single-ion anisotropy by magnon-magnon interaction. The unimportance of these higher-order contributions is confirmed by a comparison of the aniso-

ropy in the easy and the hard directions of Tb. When the direction of magnetization is changed from an easy to a hard axis the effective anisotropy at zero wave vector is only modified slightly, but the large \vec{q} -dependent anisotropy present at 4.2 K in Tb at zero field almost disappears when the direction of magnetization is changed (Fig. 4 of I).

As in I, we conclude that the effects of two-magnon-one-phonon interactions on the dispersion of the magnons are negligible at zero temperature. At finite temperatures, these effects can hardly be distinguished from those arising from normal magnon-magnon interaction. The contributions from $\mathcal{H}_{m-p}^{(1)}$ to the dispersion of the magnon energies are much more important, and we shall concentrate on these direct couplings between magnons and phonons in the following sections. Although the generalization of the static Hamiltonian to the dynamic case accounts for most of the couplings found experimentally in Tb,^{3,4} it fails to predict the observed coupling between acoustic magnons and optical phonons propagating in the c direction. The presence of this additional magnon-phonon interaction does not affect the discussion above concerning $\mathcal{H}_{m-p}^{(2)}$, but the derivation of a more complete expression for $\mathcal{H}_{m-p}^{(1)}$ demands a reformulation of the problem.

III. MAGNON-PHONON INTERACTION

The general spin interaction term introduced in I by Eq. (4), describing a coupling between the total angular moments on the sites \vec{R}_i and \vec{R}_j , depends on the relative positions of the ions, $\vec{R}_i - \vec{R}_j$. A virtual change of this distance $\delta\vec{R}(ij)$ will modify the strength of the coupling. To first order in the Cartesian components of the displacement, $\delta R_\alpha(ij)$, the coupling between the spin system and the lattice may be written

$$\mathcal{H}_{s-1} = \sum_{i \neq j} \sum_{\alpha} \sum_{lm} \sum_{l'm'} \delta R_\alpha(ij) D_{lm,\alpha}^{l'm'}(ij) \times \tilde{O}_{l,m}(J_i) \tilde{O}_{l',m'}(J_j) + \text{H.c.} \quad (11)$$

$D_{lm,\alpha}^{l'm'}(ij)$ is a phenomenological coupling parameter which is only nonzero if $l+l'$ is even, owing to time-reversal symmetry. We have neglected a possible polarization dependence of the general spin interaction, appearing as $\langle \tilde{O}_{\lambda,\mu} \rangle$ in Eq. (4) of I, because it is inessential in this context. The selection rules for the magnon-phonon interactions which are introduced by \mathcal{H}_{s-1} may be determined by a group-theoretical analysis. This has recently been performed by Cracknell¹¹ on ferromagnetic hcp metals. For the same purpose we shall here utilize a knowledge of the transformation prop-

erties of the Racah operators, and we restrict ourselves to the case of magnons propagating in the c direction of a basal-plane ferromagnet (Tb and Dy).

In the c direction of an hcp lattice, the eigenvectors of the phonons are purely longitudinal or transverse, and the transverse modes are doubly degenerate. The phonons are either purely acoustic or optical excitations (corresponding to the double-zone representation), which we shall assume to be the case also for the magnons (this assumption has no influence on the results deduced). When we consider excitations propagating in the c direction, the Hamiltonian (11) may be reduced by the symmetry operations which leave the hexagonal layers unchanged. The Cartesian 1, 2, and 3 axes are chosen to be along a , b , and c directions, respectively, and we define the components of the displacement vector $\delta R_\alpha(ij)$ to be real quantities. Referring to a coordinate system of the spin system whose axes coincide with the Cartesian axes, the general spin-lattice Hamiltonian is reduced according to the following rules: The interaction between the magnons and the longitudinal phonons is determined by

$$m+m'=3p, \quad (12a)$$

$$D_{lm,i}^{l'm'}(ij) = D_{lm,3}^{l'm'}(ij) = (-1)^p D_{lm,i}^{l'm'}(ij)^*,$$

and the interaction with the transverse phonons by

$$m+m'+1=3p, \quad (12b)$$

$$D_{lm,t}^{l'm'}(ij) = D_{lm,1}^{l'm'}(ij) = -i D_{lm,2}^{l'm'}(ij) = (-1)^p D_{lm,t}^{l'm'}(ij)^*$$

where p is equal to 0, ± 1 , ± 2 , \dots . The Hamiltonian (11) is invariant under lattice translations when

$$D_{lm,s}^{l'm'}(ij') = -(-1)^p D_{lm,s}^{l'm'}(ij), \quad (13a)$$

where $(ij) \rightarrow (ij')$ by a reflection in the hexagonal plane which includes the i th ion (the index s stands for either l or t). The transformation of one sublattice (A) into the other sublattice (B) by which $(i_A j) \rightarrow (i_B j)$ implies that

$$D_{lm,s}^{l'm'}(i_B j) = (-1)^p D_{lm,s}^{l'm'}(i_A j). \quad (13b)$$

Like the conditions of I for the spin-wave Hamiltonian, conditions (12) and (13) require that the ordered moments be parallel and of equal magnitude within any particular hexagonal layer.

The expansion of the Racah operators, as deduced in I [Eqs. (16) and (17)] for a basal-plane ferromagnet, is introduced in (11). The presence of two ions per unit cell is accounted for by defining two systems of spin deviation operators, each associated with one of the two sublattices.¹²

The relative displacement $\delta R_\alpha(ij)$ is expressed in terms of phonon operators. Introducing the conditions (12) into the spin-lattice Hamiltonian, we deduce the selection rules for the linear coupling between magnons and phonons propagating in the c direction of a basal-plane ferromagnet. Defining ψ_s as the angle between the magnetization vector and the polarization vector of the phonon mode s under consideration, the magnon-phonon interaction at a certain wavelength is proportional to

$$\sum_p D_{s,f}^p [1 + (-1)^p \delta_{s,f}] \cos(3p\phi + \psi_s), \quad (14)$$

where ϕ is the angle the magnetization makes with the 1 axis. The index f defines the magnon mode to be either acoustic or optical, and $\delta_{s,f}$ is 1 if the modes are both acoustic or optical, otherwise $\delta_{s,f}$ is -1 .

The selection rules become more comprehensible if we distinguish between the two cases:

(a) p even. In this case the longitudinal phonons ($\psi_i = \frac{1}{2}\pi$) may only interact with the magnons if the magnetization is not along a symmetry direction and if $D_{i,f}^p \neq 0$ when $p \neq 0$ [corresponding to $B_{66}^{(i)}$ in Eq. (6) of III]. If $3p\phi$ is a multiple of π (the magnetization is then along an a or a b axis), only the transverse phonons for which the polarization vector is parallel to the magnetization interact with the magnons. These terms only give rise to magnon-phonon interaction if the modes are both acoustic or optical, and the interaction amplitude is proportional to k in the long-wavelength limit. These couplings are the same as those obtained by the generalization of the static Hamiltonian, Eq. (6) of III, and are effectively included in the interaction Hamiltonian (3).

(b) p odd. The coupling introduced by these terms is only nonzero if one of the modes is an acoustic and the other an optical mode. We point out that the conditions (13) imply that the terms for which i and j in (11) belong to the same sublattice are antisymmetric in spin space [see mechanism (ii) in I], whereas all other couplings are symmetric. In an a -axis magnet only the transverse phonons which are polarized parallel to the magnetization are coupled to the magnons. When the magnetization is along a b axis ($3p\phi = \pm \frac{1}{2}\pi + 2n\pi$), then the polarization vector of the phonons which are coupled to the magnons is perpendicular to the magnetization. The coupling for which p is odd is proportional to k^2 in the long-wavelength limit, and this coupling does not appear in the static Hamiltonian or in the interaction Hamiltonian (3).

This coupling scheme, expressed through the relation (14), is identical to the one obtained by Cracknell¹¹ by a group-theoretical analysis, and it

is summarized in Table I. The coupling for which p is odd introduces interactions between acoustic and optical modes which could account for the appearance of the energy gap Δ_2 in the magnon dispersion relation in the c direction of Tb shown in Fig. 1. However, the experimental studies of the magnon-phonon interactions in Tb, which are presented later, showed no indications of such a b coupling. The acoustic-optical magnon-phonon interaction in Tb cannot be described as a b coupling, which shows that Tb is not an ideal basal-plane ferromagnet. The ionic angular moments in Tb seem to constitute a well-defined spin-wave system at low temperatures. The presence of magnetic anisotropy affects the ground state, and Lindgård and Danielsen¹⁰ found that the moments are reduced by 0.6%. A deviation of the ground state from the fully aligned spin-wave state does not introduce new selection rules if the magnetic excitations are pure dipole transitions ($\Delta M_J = \pm 1$), as in the case considered by Lindgård and Danielsen. If the magnetic excitations in Tb are mixed excitations for which ΔM_J is both ± 1 and ± 2 then the selection rules above are no longer valid. The presence of such a mixing presupposes a canting of the ionic moments in the ground state; the actual value of ϕ should deviate somewhat from the one obtained in a spin-wave approach. The effect of a canting of the ionic moments is simply accounted for by introducing the actual value of ϕ in Eq. (14). The magnetization measurements¹³ and the neutron-cross-section calculations in I show that the canted moment is at least one order of magnitude smaller than the total moment, which

TABLE I. Selection rules for the linear coupling between acoustic and optical magnons (MA, MO) and phonons propagating in the c direction of a ferromagnetic hcp metal. The moments are ordered along either an a axis or a b axis. TA_{\parallel} and TA_{\perp} label the transverse (acoustic) phonons which have their polarization vector parallel or perpendicular to the magnetization, respectively. a and b classify the different couplings which may occur, as described in the text. The spin-mixed-induced couplings introduced by Eq. (15) may appear in all those places which are not classified as an a or a b coupling.

| | a -axis magnet | | b -axis magnet | |
|------------------|------------------|-----|------------------|-----|
| | MA | MO | MA | MO |
| LA | ... | ... | ... | b |
| TA_{\parallel} | a | b | a | ... |
| TA_{\perp} | ... | ... | ... | b |
| LO | ... | ... | b | ... |
| TO_{\parallel} | b | a | ... | a |
| TO_{\perp} | ... | ... | b | ... |

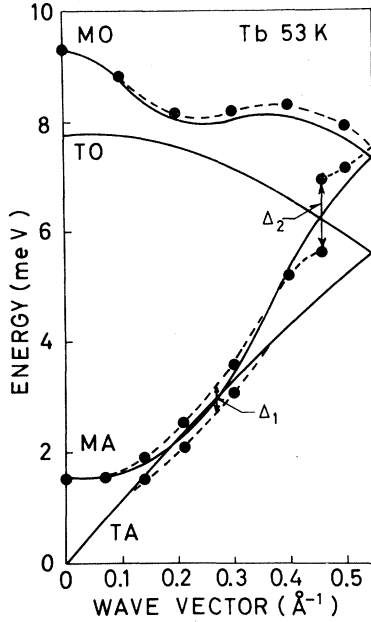


FIG. 1. Acoustic- and optical-magnon (MA, MO) and transverse-phonon (TA, TO) dispersion relations in the c direction of Tb in zero magnetic field at 53 K (solid lines). The magnon dispersion relation is deduced from the observed energies of the normal modes, connected by the dashed lines, as described in the text (the scattering vector of the neutrons is along the c axis). The normal modes are mixed states of magnons and phonons and the energy gaps Δ_1 and Δ_2 occur at the crossing points of the unperturbed dispersion relations.

implies that the $\Delta M_J = \pm 2$ transitions can be neglected. The deviation of the ground state of the ionic moments in Tb from an aligned spin-wave state is so small that it offers no possibilities for explaining the behavior of the strong acoustic-optical magnon-phonon interaction.

Until now we have considered the system of the phonons and the ionic angular moments to be an isolated one. However, the conduction electrons are known to be a very important part of the magnetic system in the rare-earth metals since they are responsible for the strong coupling between the ionic moments on different sites (I). The transformation of the s - f exchange Hamiltonian to an effective f - f Hamiltonian, by which the angular-moment space is uncoupled from the spin space of the conduction electrons, is in general a fair approximation. When making this transformation it is normally assumed that the conduction electrons are polarized parallel to the ionic moments and hence contribute to the total magnetization (4% in Tb). This condition is not necessarily fulfilled if the conduction electrons are spin-orbit coupled, in which case the spin-up and the spin-down states may be mixed. A mixing of the spin states is

equivalent to a deviation between the direction in which the conduction electrons are polarized and the direction of the magnetization. The spin component perpendicular to the magnetization is found to be proportional to a spin-orbit coupling parameter $\langle \lambda_{s-o} \rangle$, the s - f exchange matrix element, and the susceptibility of the conduction electrons. The microscopic mechanism proposed by Liu² for explaining the acoustic-optical magnon-phonon interaction involves a perpendicular spin component created by the spin-orbit coupling. The magnon-phonon interaction considered by Liu is established via intermediate states in which electron-hole pairs are excited virtually by the phonons. These pairs subsequently recombine into magnons, and this interaction becomes possible only if the spin states are mixed by the spin-orbit coupling. As estimated by Liu² a spin mixing parameter $\langle \lambda_{s-o} \rangle$ of the order of 0.03 is sufficiently large to account for the observed strengths of the acoustic-optical magnon-phonon interactions in Tb and Dy.¹⁴ The interactions due to this coupling mechanism do not appear in the selection rules above, (14), because they are proportional to the spin mixing or to the spin component of the conduction electrons perpendicular to the magnetization.

The selection rules deduced are only valid if the spin states of the conduction electrons are pure spin-up or spin-down states. The occurrence of a perpendicular spin component (parallel or perpendicular to the c axis) introduces different selection rules. From the spin-lattice Hamiltonian, (11)–(13), we deduce the following expression determining the magnon-phonon interactions which may be present in a basal plane ferromagnet to first order in the spin mixing parameter:

$$\langle \lambda_{s-o} \rangle \sum_p D_{s,f}^p [1 + (-1)^p \delta_{s,f}] \sin(3p\phi + \psi_s), \quad (15)$$

where we have used the same notation as in (14). The couplings introduced by the spin-mixing mechanism proposed by Liu² are all those which are not allowed in a simple ferromagnet. The effects of a perpendicular spin component on the spin Hamiltonian [Eqs. (7) and (8) of I] are in most cases negligible. It gives rise to couplings proportional to $\sin(\mu + m + m')\phi$ which are zero when the magnetization is along an a or a b axis, except for the interaction between acoustic and optical magnons (proportional to $\sin 3\phi$).

The acoustic-optical couplings (p odd) may only be present if the spin and space variables are directly mixed. This requirement is only met by the normal b coupling, (14), to second order in the spin-orbit parameter $\langle \lambda_{s-o} \rangle$. The absence of the b coupling in Tb is then consistent with the

spin-mixing model. The violation of the selection rules valid for a simple basal-plane ferromagnet, (14), by the acoustic-optical magnon-phonon interaction in Tb proves that this coupling is due to the spin-mixing mechanism proposed by Liu.²

The spin-mixed-induced magnon-phonon interactions, (15), may be classified according to whether p is even or odd, as in the case of the normal couplings. Yafet¹⁵ has shown that the matrix element for the phonon-electron interaction producing a spin reversal is proportional to k^2 (or k^3) in the long-wavelength limit. This implies that the couplings in (15) for which p is even are proportional to k^3 rather than k in this limit, and they do not then appear in the static Hamiltonian. The couplings for which p is odd in (14) and (15) introduce interactions between acoustic and optical modes which are proportional to k^2 in the long-wavelength limit. This implies that they cannot give rise to static (optical) deformations of the crystal.

The selection rules (15) for the spin-mixed-induced couplings do not depend on the direction of the spin component perpendicular to the magnetization. To introduce some simplification in the following discussion we shall assume that this component is perpendicular to the c axis in Tb. According to the model proposed by Liu and the considerations above the spin-mixed-induced couplings should depend on the s - f exchange interaction, $\mathcal{J}(\vec{k})$ [Eq. (1) of I], and we shall make use of the following expression for the amplitude of the acoustic-optical magnon-phonon interaction, which probably includes the most important \vec{k} -dependent contributions:

$$V_f(k) = V_0 [J \mathcal{J}_f(k) \chi_f(k)]^{1/2} \left(\frac{\epsilon_f(k)}{\omega_g(k) [A_f(k) - B_f(k)]} \right)^{1/2} \times [1 - \cos(kc)] . \quad (16)$$

The acoustic and optical branches are labeled by 1 and 2, respectively. $V_f(k)$ is the amplitude of the interaction between the magnons (f branch) and the transverse phonons (g branch) propagating in the c direction of a basal-plane ferromagnet when (f, g) is either (1, 2) or (2, 1). V_0 is a constant independent of the wave vector, and $\chi_f(k)$ is the susceptibility of the conduction electrons.¹⁶

Before finishing this section we shall consider briefly other kinds of magnetically ordered systems. If the magnetization is along the c axis then the normal b coupling vanishes, but the a coupling between the transverse phonons and the magnons may still be present [$m = -1$ and $m' = 0$ corresponding to $p = 0$ in (12b)]. If the spin states are mixed there might also be a coupling between acoustic and optical modes ($m = m' = 1$) in this configuration.

In a helically or conically ordered structure the spin-lattice Hamiltonian (11) introduces (first-order) couplings between a magnon at wave vector \vec{q} and a phonon at wave vector $\vec{q} \pm (m+m')\vec{Q} + n\vec{\tau}_c$, where \vec{Q} is the wave vector of the magnetic structure, $|\vec{\tau}_c| = 2\pi/c$, and n is an integer which is even/odd when p is even/odd. These are the selection rules obtained when using a double-zone representation for the modes propagating in the c direction, and they are valid both for the normal and spin-mixed-induced couplings. A coupling between the transverse phonon modes at $\vec{q} = 0.17\vec{\tau}_c$ and the magnon at $\vec{q} = -0.35\vec{\tau}_c$ for which $m+m' = 2$ and p is odd (corresponding to the acoustic-optical interaction in Tb) is presumably responsible for the energy gap at $\vec{q} = -0.35\vec{\tau}_c$ observed in the spin-wave spectrum of Er ($\vec{Q} = 0.24\vec{\tau}_c$) by Nicklow *et al.*¹⁷

IV. EQUATIONS OF MOTION

In this section we shall consider the equations of motion of the coupled magnon-phonon system, and we shall neglect the couplings which seem to be of no importance in Tb. This means that we restrict ourselves to the case where the magnons and the transverse phonons propagating in the c direction are coupled via the a coupling (see Table I) and the acoustic-optical spin-mixed-induced coupling, (15). The selection rules for the acoustic-optical interaction make it necessary to distinguish between the two cases in which the magnetization is either along an a direction or along a b direction. If we distinguish between the transverse phonons polarized parallel ($\beta_{\vec{k}}$) and perpendicular ($\gamma_{\vec{k}}$) to the magnetization by defining two sets of phonon operators, $\beta_{\vec{k}}$ and $\gamma_{\vec{k}}$, then the unperturbed Hamiltonian for the magnon-transverse-phonon system is

$$\mathcal{H}_{m-\text{tp}}^0 = \sum_{\vec{k}} \sum_{f=1,2} [\epsilon_f(\vec{k}) \alpha_{f,\vec{k}}^\dagger \alpha_{f,\vec{k}} + \omega_f(\vec{k}) \beta_{f,\vec{k}}^\dagger \beta_{f,\vec{k}} + \omega_f(\vec{k}) \gamma_{f,\vec{k}}^\dagger \gamma_{f,\vec{k}}] . \quad (17)$$

When the magnetization is along an a axis these modes are coupled as described by the Hamiltonian

$$\mathcal{H}_{m-\text{tp}}^a = \sum_{\vec{k}} \sum_{f=1,2} [iW_f(\vec{k}) (\alpha_{f,\vec{k}}^\dagger + \alpha_{f,-\vec{k}}) (\beta_{f,\vec{k}} + \beta_{f,-\vec{k}}^\dagger) + iV_f(\vec{k}) (\alpha_{f,\vec{k}}^\dagger - \alpha_{f,-\vec{k}}) (\gamma_{g,\vec{k}} + \gamma_{g,-\vec{k}}^\dagger)] , \quad (18a)$$

which is changed into

$$\mathcal{H}_{m-\text{tp}}^b = \sum_{\vec{k}} \sum_{f=1,2} [iW_f(\vec{k}) (\alpha_{f,\vec{k}}^\dagger + \alpha_{f,-\vec{k}}) (\beta_{f,\vec{k}} + \beta_{f,-\vec{k}}^\dagger) + iV_f(\vec{k}) (\alpha_{f,\vec{k}}^\dagger - \alpha_{f,-\vec{k}}) (\beta_{g,\vec{k}} + \beta_{g,-\vec{k}}^\dagger)] \quad (18b)$$

when the magnetization is along a b axis. In these equations $g=2$ when $f=1$ and vice versa. $W_f(\vec{k})$ and $V_f(\vec{k})$ are both real quantities. When \vec{k} is replaced by $-\vec{k}$, then $W_f(\vec{k})$, Eq. (5), changes sign, whereas $V_f(\vec{k})$, Eq. (16), is unchanged. By expressing the Hamiltonian in terms of magnon operators, we have performed the transformation of the spin deviation operators into magnon operators [Eq. (19) of I]. This transformation affects only the interaction amplitudes [as included in (5) and (16)], not the equations of motion.

The Hamiltonian for the coupled magnon-transverse-phonon system, (17) and (18), also describes the case where the two normal (a and b) interactions are the only ones present. The only difference is that the Hamiltonian for the a -axis

magnet (18a) and the Hamiltonian for the b -axis magnet (18b) are interchanged. By accounting for this difference the following discussion of the equations of motion is equally valid in the two cases.

The normal modes of the system described by (17) and (18) are certain linear combinations of the magnon and phonon modes. By evaluating the secular determinants, we obtain the following eigenvalue equations which determine the energy $E(\vec{k})$ of the normal modes. The wave-vector argument is the same for all the quantities occurring in the following equations, and we therefore suppress it. In the case where the magnetization is along an a axis the equations of motion lead to

$$\begin{aligned} & [(E^2 - \epsilon_1^2)(E^2 - \omega_1^2)(E^2 - \omega_2^2) - 4\epsilon_1\omega_1W_1^2(E^2 - \omega_2^2) - 4\epsilon_1\omega_2V_1^2(E^2 - \omega_1^2)][(E^2 - \epsilon_2^2)(E^2 - \omega_2^2)(E^2 - \omega_1^2) \\ & - 4\epsilon_2\omega_2W_2^2(E^2 - \omega_1^2) - 4\epsilon_2\omega_1V_2^2(E^2 - \omega_2^2)] = 0, \end{aligned} \quad (19a)$$

and when $\mathcal{H}_{m-tp}^c = \mathcal{H}_{m-tp}^a + \mathcal{H}_{m-tp}^b$ we obtain

$$\begin{aligned} & (E^2 - \omega_1^2)(E^2 - \omega_2^2)[(E^2 - \epsilon_1^2)(E^2 - \epsilon_2^2)(E^2 - \omega_1^2)(E^2 - \omega_2^2) - 4\epsilon_1\omega_1W_1^2(E^2 - \epsilon_2^2)(E^2 - \omega_2^2) - 4\epsilon_2\omega_2W_2^2(E^2 - \epsilon_1^2)(E^2 - \omega_1^2) \\ & - 4\epsilon_1\omega_2V_1^2(E^2 - \epsilon_2^2)(E^2 - \omega_1^2) - 4\epsilon_2\omega_1V_2^2(E^2 - \epsilon_1^2)(E^2 - \omega_2^2) + 16\omega_1\omega_2(\epsilon_1W_2^2 + \epsilon_2V_1^2)(\epsilon_2W_1^2 + \epsilon_1V_2^2) \\ & - 16\omega_1\omega_2E^2(W_1V_1 + W_2V_2)^2] = 0. \end{aligned} \quad (19b)$$

The six positive roots E of these equations are the energies of the six (different) normal modes of the system. Putting V_1 and V_2 equal to zero in (19a) or (19b) we may easily derive Eqs. (6) and (7) in Sec. II.

At point A on the Brillouin-zone boundary the magnons and phonons are both doubly degenerate, ϵ_1 and ϵ_2 equal to ϵ and ω_1 and ω_2 equal to ω . Eq. (19a) immediately shows that the normal modes remain doubly degenerate at A when the magnetization is along an a axis. If the magnetization is parallel to a b direction, then the energies of the normal modes are determined by

$$\begin{aligned} & (E^2 - \omega^2)^2[(E^2 - \epsilon^2)(E^2 - \omega^2) - 4\epsilon\omega(W^2 + V^2) - 8\omega EWV] \\ & \times [(E^2 - \epsilon^2)(E^2 - \omega^2) - 4\epsilon\omega(W^2 + V^2) + 8\omega EWV] = 0 \end{aligned} \quad (20)$$

at $k = \pi/c$. This equation will in general have $4 + 1$ different positive roots, implying that the double degeneracy of the coupled modes at A is lifted (when W and V are both nonzero). This indirect coupling of the acoustic and the optical magnons in a b -axis magnet, which is transmitted by the combined acoustic and optical interaction with the phonons, is a higher-order process. In

a second-order perturbation calculation this coupling will not appear, and thus it does not violate the general symmetry arguments in I.

The energy gap at A obtained in the b -axis magnet, Eq. (20), is a result of the particular spin-mixed-induced coupling which we consider. If the spin component perpendicular to the magnetization is parallel to the c axis instead of lying in the basal plane, as we have assumed, then the equations above are modified in the following way. In Eq. (16), the expression for the interaction amplitude, $A_f(k) - B_f(k)$ should be replaced by $A_f(k) + B_f(k)$. In the Hamiltonian (18), $i(\alpha_{f,\vec{k}}^\dagger - \alpha_{f,-\vec{k}})$ should be changed into $\alpha_{f,\vec{k}}^\dagger + \alpha_{f,-\vec{k}}$, which modifies the eigenvalue equation (19b), so that the last two terms are replaced by $16\epsilon_1\epsilon_2\omega_1\omega_2 \times (W_1W_2 + V_1V_2)^2$. This change of the eigenvalue equation (19b) implies that, if the spin component giving rise to the spin-mixed-induced coupling is parallel to the c axis, then the magnons (and phonons) also remain doubly degenerate at A when the magnetization is along a b axis.

If $W_f(\vec{k})$ and $V_f(\vec{k})$ are different from zero, then the magnon and phonon states are mixed. From the secular equations, the eigenvector describing the normal mode with energy $E(\vec{k})$ may be deduced.

We shall reduce this problem by considering only the cases where either $W_f(\vec{k})$ or $V_f(\vec{k})$ may be neglected. If unperturbed magnon and phonon branches are sufficiently close to each other, then the interaction U between these two branches dominates the behavior of the corresponding normal modes. When the coupling to the other branches is neglected the energies are determined by

$$E_{\pm} = \left[\frac{1}{2}(\epsilon^2 + \omega^2) \pm \frac{1}{2}(\epsilon^2 - \omega^2)F \right]^{1/2},$$

$$E_{\epsilon} = E_{+} \quad \text{and} \quad E_{\omega} = E_{-}, \quad (21)$$

where F is equal to 1 if the interaction vanishes,

$$F = [1 + 16\epsilon\omega U^2 / (\epsilon^2 - \omega^2)^2]^{1/2}. \quad (22)$$

When $\epsilon \neq \omega$, the mode with the energy E_{ϵ} is mostly magnonlike and the other is mostly phononlike. The inelastic neutron scattering cross section of the transverse phonons vanishes if the scattering vector \vec{k} is along a c direction, which is in distinction to the cross section of the magnons [Eq. (43) of I]. The cross sections of the two normal modes are then entirely connected to the magnon part of their eigenvectors. By calculating these eigenvectors we obtain the ratio between the neutron intensities of the two modes, I_{ϵ} and I_{ω} , as

$$I_{\omega}/I_{\epsilon} = K_{\omega\epsilon} E_{\omega}(F-1)/E_{\epsilon}(F+1), \quad (23a)$$

where $K_{\omega\epsilon}$ is a correction due to a different occupation $n(E)$ of the two levels and to a change of the ratio $(k'/k_0)_{\epsilon}$ between the wave vectors of the incident and scattered neutrons,

$$K_{\omega\epsilon} = \frac{(n_{\omega} + 1)(k'/k_0)_{\omega}}{(n_{\epsilon} + 1)(k'/k_0)_{\epsilon}}. \quad (23b)$$

If we approximate $\omega\epsilon$ by $E_{\omega}E_{\epsilon}$ then F may be expressed in terms of the observed energies

$$F \cong |E_{\omega}^2 - E_{\epsilon}^2| / [(E_{\omega}^2 - E_{\epsilon}^2)^2 - 16U^2 E_{\omega}E_{\epsilon}]^{1/2}. \quad (24)$$

Equations (23) and (24) may be used to obtain a fair separation of overlapping neutron groups.

V. EXPERIMENTS

The interactions between magnons and phonons propagating in the c direction of Tb were studied by inelastic neutron scattering. No coupling was observed between the magnons and the longitudinal phonons. The transverse phonons and the magnons are coupled, as is revealed by the two energy gaps Δ_1 and Δ_2 in the magnon spectrum of Tb in zero field at 53 K, which is shown in Fig. 1. The coupling between the acoustic magnons and the acoustic transverse phonons Δ_1 may immediately be explained by the normal a coupling (Table I). The interaction between the acoustic magnons and the optical phonons, Δ_2 , has been proposed by

Liu² as being due to the spin-mixing mechanism. As we have seen this explanation can be verified by a determination of the polarization vector of the transverse phonon mode which takes part in the interaction. When the magnetization is along the b axis (as in Tb in zero field), then the polarization vector of the optical transverse phonons, which are coupled to acoustic magnons, should be perpendicular to the magnetization in the case of a simple ferromagnet (Table I); whereas the spin mixing mechanism, (15), predicts the polarization vector to be parallel to the magnetization. In Fig. 2, we show the result of a neutron scan at $q = 0.6 \text{ \AA}^{-1}$ at 90 K. The reciprocal-lattice vector defining this scan is $(1, 1, 0)$, in which case the neutron cross sections are finite only for optical modes when q is larger than $\pi/c = 0.55 \text{ \AA}^{-1}$. To remove domain effects we applied an external field along the b axis perpendicular to the scattering plane; these effects are small because the magnetovibrational cross section dominates the phonon creation through the nuclear interaction. In this configuration we detect the optical magnon (MO) and the optical phonon polarized perpendicular to the magnetization (TO_{\perp}) at $q = 0.5 \text{ \AA}^{-1}$. As is apparent on Fig. 2, the TO_{\perp} mode is not affected by the acoustic magnon mode (MA). If the coupling giving rise to the energy gap Δ_2 in the magnon spectrum had been of the normal b type then the neutron group of the TO_{\perp} mode would have split into two peaks appearing at the positions marked by the two arrows in Fig. 2. We did not observe any anomaly in the behavior of the neutron intensity or linewidth of the TO_{\perp} mode when q was varied, implying that the b coupling in Tb is entirely negligible. By the application of a field along an a direction the ordered moments were aligned along this direction perpendicular to the scattering plane. In this configuration it should in principle be possible to detect the spin-mixed-induced coupling in the spectrum of the transverse phonons polarized perpendicular to the magnetization, see Eq. (15). The experimental resolution was adequate for this purpose only around the reciprocal lattice point $(1, 0, 0)$, in which case the neutron cross section is not purely acoustic or optical, and it was not possible to separate the neutrons scattered by the acoustic magnons from those scattered by the optical phonons in the neighborhood of the energy gap Δ_2 . However, the energy gap appeared as two separate neutron groups in this scan, and the ratio between the neutron intensities of the optical transverse phonon and of the acoustic magnon was sufficiently large for an uncoupled phonon mode to have smeared out the two neutron groups completely. From these experiments we conclude that the

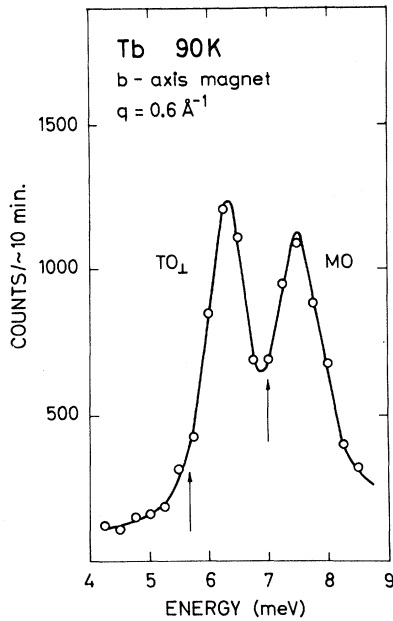


FIG. 2. Neutron groups obtained in Tb at 90 K when a field of 10 kOe is applied along a b axis perpendicular to the scattering plane. The scattering vector of the neutrons is the sum of the reciprocal lattice vector $(1, 1, 0)$ and the wave vector \vec{q} along the c axis ($q = 0.6 \text{ \AA}^{-1}$). The two peaks observed are due to the optical magnon mode (MO) and the optical transverse-phonon mode polarized perpendicular to the magnetization (TO_{\perp}) at $q = 0.5 \text{ \AA}^{-1}$. If the energy gap Δ_2 in Fig. 1 were due to the normal b coupling appearing in Table I, then the TO_{\perp} neutron group should have split into two peaks of equal magnitudes at the positions marked by the arrows.

acoustic-optical magnon-phonon interaction in Tb which is manifested as the energy gap Δ_2 is a spin-mixed-induced coupling, as proposed by Liu.²

If the spin component of the conduction electrons, which is perpendicular to the magnetization, is perpendicular to the c axis, then the magnons (and phonons) should exhibit an energy gap at the Brillouin-zone boundary (A) when the magnetization is along a b axis, as determined by Eq. (20). From the interaction amplitudes derived below we calculate the energy gap at A to be approximately 0.28 meV at 4.2 K. This is relatively small when compared with the natural linewidth of the magnon¹⁸ at A , which is of the order of 0.6 meV. The intensity variation of the (perturbed) magnons was studied in the vicinity of the Brillouin-zone boundary. The experimental resolution was 0.5 meV, corresponding to an observed linewidth of the order 0.8 meV. The experiment suggested an energy gap equal to 0.25 ± 0.15 meV, but this is too small to allow a direct verification of

its existence. The uncertainty introduced in the eigenvalue equation (19b) because the direction of the perpendicular spin component is unknown is of minor importance except at A , and we shall continue to assume that this component lies in the basal plane.

The magnitude of the energy gaps which occur at the nominal crossing points of the magnon and phonon dispersion relations is approximately twice the amplitude of the magnon-phonon coupling, Eq. (6). The use of the exact expression (19) produces only a small correction. By measuring the energy gaps we may determine the strength of the couplings at the wave vectors at which the energy gaps appear. From the energy gaps $\Delta_1(\vec{k}_1)$ and $\Delta_2(\vec{k}_2)$, which are shown in Fig. 1, we deduce the coupling amplitudes $W_1(\vec{k}_1)$ and $V_1(\vec{k}_2)$. By the application of an external magnetic field the ordered moments were aligned along a hard direction (a axis). From the magnitudes of the energy gaps observed in the two different configurations the coupling parameters H_0 and V_0 , Eqs. (5) and (16), were found to be independent of ϕ . Away from an energy gap the intensity ratio (23) decreases rapidly and may serve only as an order-of-magnitude estimate of the coupling strength. The interaction between acoustic magnons and optical phonons at zero wave vector, $V_1(0)$, does not necessarily vanish as suggested by Eq. (16). The intensity of the optical transverse phonon mode at $q = 0$, I_{ω} in Eq. (23), was found to be vanishingly small when the scattering vector of the neutrons was parallel to the c axis. From this measurement an upper limit of 0.1 meV could be deduced for $|V_1(0)|$, which is consistent with the factor $1 - \cos(kc)$ in (16).

We did not find any indications in Tb of a spin-mixed-induced coupling for which p in (15) is even. The presence of such a coupling might be of importance only close to A because it is proportional to $(kc)^3$ rather than kc . Neglecting this coupling and a possible deviation of the order of $(kc)^3$ which may occur in the expression for $W_f(k)$, Eq. (5), we calculated the intensity ratio (23) between the neutrons scattered by the transverse phonon and the magnon at A . The experimental value was found to be close to the calculated one, supporting the use of these approximations. Within these approximations the result for $W_1(\vec{k}_1)$ at 53 K determines H_0 as 17.0×10^{-3} at $\sigma = 0.971$ by the use of Eq. (5). The velocity of acoustic sound waves depends on an applied field, Eq. (7), because of the field dependence of the magnon energy gap at zero wave vector (III). The acoustic-optical coupling does not contribute in this long-wavelength limit. Moran and Lüthi¹⁹ have measured the field dependence of the velocity of transverse sound

waves propagating in the c direction of Tb at 140 K from which H_0 at $\sigma = 0.82$ is deduced³ to be 14.6×10^{-3} [this number has been corrected for the effect of the field dependence of the relative magnetization (III)]. The magnetostriction parameter $\lambda^\epsilon = \frac{1}{2}H_0$ has been measured^{20,21} only in the paramagnetic region of Tb ($\sigma < 0.2$). The combination of these measurements with the two results deduced from the magnon-phonon interaction indicates (Fig. 3) a simple power-law dependence on σ of H_0 instead of the $\hat{I}_{5/2}(\sigma)$ dependence obtained from the theory of Callen and Callen¹ when H_0 is assumed to be of lowest-order single-ion origin. By fitting a power-law σ dependence to λ^ϵ measured by DeSavage and Clark²⁰ and to the two results deduced at low temperatures we obtain $H_0 = 2\lambda^\epsilon = 18.5 \times 10^{-3} \times \sigma^{1.76}$, as shown in Fig. 3.

This value for H_0 has been used in (5) to calculate $W_f(k)$. Defining

$$\begin{aligned} W(k) &= W_1(k), \quad V(k) = V_1(k), \\ \text{when } 0 < k < \pi/c, \\ W(k) &= W_2(2\pi/c - k), \quad V(k) = V_2(2\pi/c - k), \\ \text{when } \pi/c < k < 2\pi/c, \end{aligned} \quad (25)$$

corresponding to the double-zone representation, we show in Fig. 4 $W(k)$ as a function of k in Tb at 4.2 K. The magnetization is along an easy axis. The experimental result shown at $k_1 = 0.3 \text{ \AA}^{-1}$ was obtained from the measurements at 53 K (the phonon and magnon branches do not cross at 4.2 K). To obtain $W(k_1)$ at $\sigma = 1$ we have modified the result at $\sigma = 0.971$ according to Eq. (5), where H_0 depends on σ as given above and where c_ϵ implicitly includes a factor $1/\sigma$ (see III). Determining V_0 from the energy gap at 4.2 K, $\Delta_2 = 1.30 \text{ meV}$ at $k_2 = 0.45 \text{ \AA}^{-1}$, we have deduced $V(k)$ shown in Fig. 4 from the predicted wave-vector dependence, Eq. (16). $J\mathcal{G}(k)$ was replaced by $J\mathcal{G}(0) - g(k)$, where $g(k)$ is defined in I [Eqs. (39)–(42)] and $J\mathcal{G}(0) = 8.0 \text{ meV}$. Further we have used the susceptibility $\chi(k)$ calculated by Liu *et al.*¹⁶

Besides a knowledge of the interaction amplitudes $W_f(\vec{k})$ and $V_f(\vec{k})$, a determination of $\epsilon_f(\vec{k})$ from the perturbed magnon energy $E_f(\vec{k})$, Eq. (19), also requires a knowledge of the positions of the other magnon and phonon branches. The dispersion relation of the unperturbed transverse phonons, as measured at room temperature by Houmann and Nicklow,²² is shown in Fig. 1, and in the double zone it is quite well described by the sinusoidal k dependence, $\omega(k) = \omega_0 \sin(kc/4)$. The constant ω_0 depends slightly on the temperature, corresponding to the temperature dependence of the elastic constant c_{44} . With $\epsilon_f(\vec{k})$ as the only unknown quantity Eq. (19) may easily be solved, and by

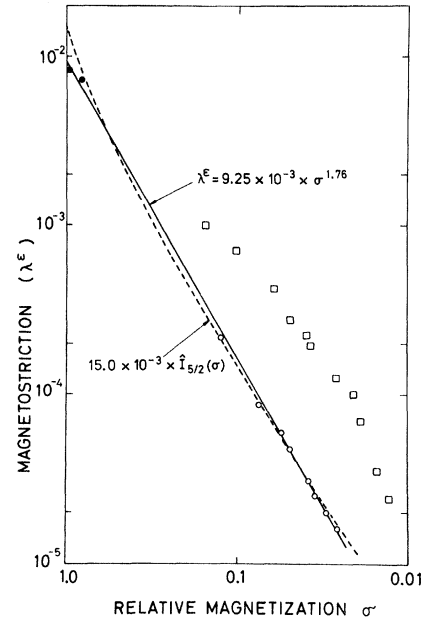


FIG. 3. ϵ -strain magnetostriction parameter $\lambda^\epsilon = \frac{1}{2}H_0$ in Tb as a function of the relative magnetization σ . The experimental results for σ smaller than 0.2 are obtained from magnetostriction experiments (O, Ref. 20, and \square , Ref. 21). The two results at low temperatures are deduced from the amplitude of the magnon-phonon interaction as described in the text. The solid line displays a simple power-law fit to the low-temperature results and to the results from Ref. 20. The dashed line shows the extrapolated behavior of the magnetostriction results (Ref. 20) based on the theory of Callen and Callen (Ref. 1), assuming λ^ϵ to be of lowest-order single-ion origin.

doing this we have obtained the dispersion relation of the unperturbed magnons propagating in the c direction of Tb at 53 K, as shown in Fig. 1.

In the interpretation of the dependence of the magnon energies in Tb on magnetic field and temperature in I, the magnon energies were corrected for the effect of magnon-phonon interaction. The corrections were performed using the values for H_0 and V_0 determined in the present paper together with Eqs. (5), (16), and (19). V_0 was assumed to be independent of temperature, in accordance with the discussion by Liu.² If $W_f(\vec{k})$ and $V_f(\vec{k})$ are assumed to be smooth functions of \vec{k} , the knowledge of the functions at a single \vec{k} value, combined with the symmetry restrictions imposed on the functions, is sufficient to obtain a satisfactory account of the effect of the magnon-phonon interaction. The \vec{k} dependences of $W_f(\vec{k})$ and $V_f(\vec{k})$ are determined only roughly by Eqs. (5) and (16), but the most important effects of these couplings occur at \vec{k} values close to \vec{k}_1 and \vec{k}_2 , where the strength of the couplings are well known. This is also the

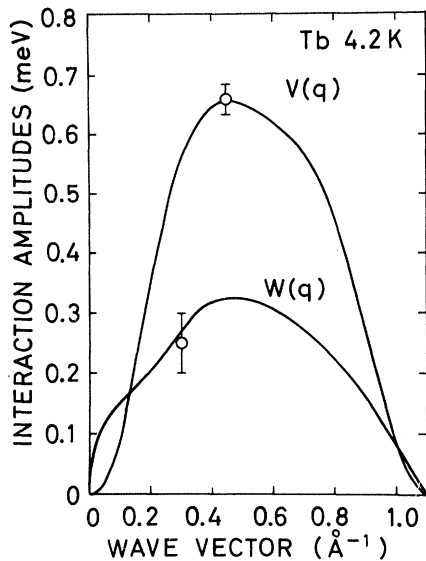


FIG. 4. Amplitudes of the magnon-phonon interactions as a function of wave vector in the c direction of Tb at 4.2 K (the magnetization is along an easy axis). The experimental results are deduced from the energy gaps Δ_1 and Δ_2 shown in Fig. 1. The solid lines show the wave-vector dependences predicted by Eqs. (5) and (16), where we have used a double-zone representation, as defined by Eq. (25).

argument through which the possible contributions of other couplings can be neglected.

VI. CONCLUSION

From general symmetry considerations, we have deduced the selection rules for the linear coupling between magnons and phonons propagating in the

c direction of a simple ferromagnetic hcp metal in which the ordered moments lie in the basal plane. The interaction mechanism proposed by Liu² for explaining the acoustic-optical magnon-phonon interaction in the rare-earth metals is found to introduce couplings which violate the selection rules valid for a simple ferromagnet. The different selection rules applicable to these couplings are a result of their dependence on a spin mixing of the conduction electrons due to the spin-orbit coupling. The interaction $V_f(\vec{k})$ between acoustic (optical) magnons and optical (acoustic) transverse phonons propagating in the c direction of Tb was observed to be a spin-mixed-induced coupling, as proposed by Liu.² This coupling appears to be about twice as big as the normal coupling $W_f(\vec{k})$ between acoustic (optical) magnons and acoustic (optical) transverse phonons. $V_f(\vec{k})$ and $W_f(\vec{k})$ are the only magnon-phonon interactions observed in the c direction of Tb, and the measured energy splittings of the unperturbed magnon and phonon dispersion relations yield sufficient information for correcting the measured magnon energies for the effect of these couplings.

The spin-mixed-induced coupling $V_f(\vec{k})$ is the largest coupling between magnons and phonons which has been observed in Tb.^{3,4} The origin of a similar coupling in Dy¹⁴ and in Er is presumably the same as in Tb. Magnon-phonon interactions are closely connected to the anisotropy of the spin system itself, and the presence of large spin-mixed-induced magnon-phonon interactions in the heavy-rare-earth metals indicates the importance of the spin-orbit coupling of the conduction electrons for the two-ion anisotropy observed in these metals (I).

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