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MAGNETO-ELASTIC INTERACTIONS IN TERBIUM

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Making use of the Hamiltonian for linear magneto-elastic coupling which has been proposed by Callen and Callen, we have deduced expressions for changes in the velocity of acoustic waves in a Terbium crystal, due to ferromagnetic ordering and the application of an external magnetic field.

These calculations agree semi-quantitatively with the results of experimental measurements. We have also examined the extent to which this simple picture is applicable to explain the magnon–phonon interactions in Terbium, which have been observed at finite wave vectors by inelastic neutron scattering.

1. INTRODUCTION

Because of their large orbital moments, the heavy rare earth metals display the largest known magnetostriction effects.[1,2] This strong coupling between the magnetic moments and the lattice has the effect that the elastic constants and the sound-wave velocities depend relatively strongly on the magnetization of the crystal. A number of experimental observations of this effect have recently been published.[3–7]. In this paper we derive expressions for the change in sound velocities below the Curie temperature due to the linear magneto-elastic interaction, in terms of the magnetostriction coefficients and the magnon energies. The change in velocity due to an applied field is also considered. The extent to which these considerations apply to the magnon–phonon interactions, which have been observed at finite wave vector by neutron scattering, is also discussed.

2. THE MAGNETO-ELASTIC HAMILTONIAN

In the spin wave theory[8,9] it is normally assumed that the ions remain in their equilibrium positions. However, the R-dependence of the exchange and crystal-field parameters causes a coupling between magnons and phonons, leading to a modification of their energy and to magnetostriction. Mason[10] has derived macroscopic expressions for the magnetostriction, while Callen and Callen[11] have developed a phenomenological theory of the magneto-elastic coupling. Following them, we write the interaction between the lattice and the spin system:

$$\begin{aligned}\tilde{H}_{S-L} = &- \sum_{ij,\alpha}[(\partial J_{ij}/\partial R_{i\alpha})\delta R_{i\alpha} \\ &+ (\partial J_{ij}/\partial R_{j\alpha})\delta R_{j\alpha}]\bar{J}_i\cdot\bar{J}_j \\ &+ G_{01}\sum_j[J_\zeta^2 - \tfrac{1}{3}J(J+1)]_j(e_{11}+e_{22}) \\ &+ G_{03}\sum_j[J_\zeta^2 - \tfrac{1}{3}J(J+1)]_j e_{33} \\ &- G_{22}\sum_j[\tfrac{1}{2}[(J^+)^2 + (J^-)^2]_j(e_{11}-e_{22}) \\ &+ (1/2i)[(J^+)^2 - (J^-)^2]_j e_{12}] \\ &- G_{13}\sum_j[\tfrac{1}{2}[J_\zeta J_\xi + J_\xi J_\zeta]_j e_{13} \\ &+ \tfrac{1}{2}[J_\zeta J_\eta + J_\eta J_\zeta]_j e_{23}] + G_{44}\sum_j[\tfrac{1}{2}[(J^+)^4 \\ &+ (J^-)^4]_j(e_{11}-e_{22}) \\ &- (1/2i)[(J^+)^4 - (J^-)^4]_j e_{12}]\end{aligned} \tag{1}$$

The strains $e_{\alpha\beta}$ are defined in terms of the elastic displacements $u_\alpha(\bar{r}, t)$:

$$e_{\alpha\beta} = (1 - \tfrac{1}{2}\delta_{\alpha\beta})(\partial u_\alpha/\partial x_\beta + \partial u_\beta/\partial x_\alpha) \tag{2}$$

In accordance with Rhyne and Legvold,[1] the coordinate system used for the hcp Tb structure is orthogonal, with the 1-(orξ-) axis along $a(11\bar{2}0)$ crystal axis, the 2- (η-) axis along the b $(10\bar{1}0)$ direction, and the 3- (ζ-) axis along the c (0001) crystal axis. The angle φ is defined as the angle between the ξ-axis (hard direction) and the z-axis. z is the direction of the net magnetization, assumed to lie in the basal plane.

$\tilde{H}_{S-L}$ has the symmetry of the hexagonal lattice. The only two-ion term which is included is the one arising from the exchange energy ($J(\bar{R}_i, \bar{R}_j)$ is assumed to be a function of $\bar{R}_i - \bar{R}_j$ only). The G_{01}, G_{03}, G_{22} and G_{13} terms comprise the total one-ion magneto-elastic Hamiltonian to the second power in the spin operators. Besides these, we have included one term of fourth power (G_{44}), which has about the same magnitude as G_{22} in Terbium. The assumption that the terms have purely one-ion character is in accordance with the measurements performed by Rhyne and Legvold.[1]

Comparing the average value of this Hamiltonian at $T = 0°\text{K}$ with Mason's magnetostriction equation,[1, 10] we obtain:

$$D_{22} = G_{22}(J - \tfrac{1}{2}) = 2Cc_{66}/NJ \qquad (3)$$

$$D_{44} = G_{44}(J - \tfrac{1}{2})(J - 1)(J - \tfrac{3}{2}) = Ac_{66}/NJ \qquad (4)$$

$$G_{13} = H_0 c_{44}/[NJ(J - \tfrac{1}{2})] \qquad (5)$$

and defining

$$D' = D + (A/2), \quad D_{01} = G_{01}(J - \tfrac{1}{2}), \quad D_{03} = G_{03}(J - \tfrac{1}{2}) \qquad (6)$$

we have

$$\partial E_{ex}/\partial e_{11} - \tfrac{1}{3}NJD_{01} = -[2D'(c_{11} - c_{66}) + Gc_{13}] \qquad (7)$$

$$\partial E_{ex}/\partial e_{23} - \tfrac{1}{3}NJD_{03} = -[2D'c_{13} + Gc_{33}] \qquad (8)$$

where

$$\partial E_{ex}/\partial e_{11} \cong \partial E_{ex}/\partial e_{33} \cong -2NJ^2(J_0 + J'_0) = 2E_{ex} \qquad (9)$$

or

$$\tfrac{1}{3}[2\partial E_{ex}/\partial e_{11} + \partial E_{ex}/\partial e_{33}] = V\partial E_{ex}/\partial V = -(E_{ex}B/T_c)\partial T_c/\partial p \qquad (10)$$

A, C, D, G and H_0 are the magnetostriction constants, defined by Mason.[10] c_{ij} are the elastic constants, $c_{66} = \frac{1}{2}(c_{11} - c_{12})$, B is the bulk modulus. N is the number of atoms in the crystal (or two times the number of primitive cells).

The Hamiltonian $\tilde{H}_{S-L}$ is mainly used for discussing magnetostriction. It can however be extended to account for the dynamical interactions between magnons and phonons. The relative displacement of the ions is allowed to depend on the position of the ions. This deviation from the homogeneous strain is then expanded in normal phonon-coordinates. After introducing the spin wave operators as defined in ref. 9, we find a Hamiltonian with the form:

$$\tilde{H} = \sum_q \varepsilon(q)(\alpha_q^+\alpha_q + \tfrac{1}{2}) + \tilde{H}^{(1)}_{S-L} + \tilde{H}^{(2)}_{S-L} + \sum_k \hbar\omega(k)(\beta_k^+\beta_k + \tfrac{1}{2}) \qquad (11)$$

where

$$\tilde{H}^{(1)}_{S-L} = \sum_k W(k)(\alpha_k^+ + \alpha_{-k})(\beta_k + \beta_{-k}^+) \qquad (12)$$

and

$$\tilde{H}^{(2)}_{S-L} = \sum_{k,q} [U(k,q)\alpha_{q+k}^+\alpha_q + V(k,q)\tfrac{1}{2}(\alpha_{q+k}^+\alpha_{-q}^+ + \alpha_q\alpha_{-q-k})] \cdot (\beta_k + \beta_{-k}^+) \qquad (13)$$

β_k and α_q are respectively phonon and magnon annihilation operators.

3. ACOUSTIC VELOCITIES

The Hamiltonian with only the first kind of interaction, $\tilde{H}^{(1)}_{S-L}$, (the effects of $\tilde{H}^{(1)}_{S-L}$ and of $\tilde{H}^{(2)}_{S-L}$ are assumed to be additive) can be diagonalized exactly by a series of canonical transformations. It causes a splitting apart of the magnon and the phonon dispersion curves, where they would have crossed in the absence of the interaction, by an amount $2|W(k)|$. The energy shift of phonons with long wavelengths is:

$$\Delta\hbar\omega(k) = -2|W(k)|^2/\varepsilon(0) \qquad (14)$$

when $\varepsilon(0) \neq 0$, as is the case in Terbium.

The Hamiltonian involving the other kind of interaction, $\tilde{H}^{(2)}_{S-L}$, can be diagonalized approximately by the equation of motion method. New phonon and magnon operators are constructed, defined by

$$\gamma_k = \beta_k + \delta\beta_k - \tfrac{1}{2}[\delta\beta_k, \delta\beta_k^+]\beta_k \qquad (15)$$

where

$$\delta\beta_k = \sum_q \frac{U(k,q)}{\varepsilon(q+k) - \varepsilon(q) - \hbar\omega(k)}\alpha_q^+\alpha_{q+k} + \frac{1}{2}\frac{V(k,q)}{\varepsilon(q+k) + \varepsilon(q) - \hbar\omega(k)}\alpha_{q+k}\alpha_{-q} - \frac{1}{2}\frac{V(k,q)}{\varepsilon(q+k) + \varepsilon(q) + \hbar\omega(k)}\alpha_q^+\alpha_{-q-k}^+ \qquad (16)$$

We then have:

$$[[\gamma_k, \tilde{H}], \gamma_k^+] = \hbar\omega(k) + \Delta\hbar\omega(k) = \hbar\omega(k) + \langle[\delta\beta_k, [\tilde{H}^{(2)}_{S-L}, \beta_k^+]]\rangle \quad (17)$$

where all terms which are not diagonal in magnon and phonon operators are neglected. The energies of phonons with long wavelengths are then changed by an amount:

$$\Delta\hbar\omega(k) = \sum_q \left[|U(k,q)|^2 \cdot \frac{\partial n(q)}{\partial\varepsilon(q)} - |V(k,q)|^2 \cdot \frac{n(q)}{\varepsilon(q)} \right] \quad (18)$$

where

$$n(q) = \frac{1}{\exp[\varepsilon(q)/k_B T] - 1} \quad (19)$$

The resulting changes in velocities due to the linear magneto-elastic coupling are:

$$\Delta v_\alpha / v_\alpha = -\frac{2NJ}{c_{\alpha\alpha}(A_1(0) - B)} \Gamma_\alpha^2 + \frac{1}{2c_{\alpha\alpha}} \sum_{q,i} \frac{1}{\varepsilon_i(q)^2} \left\{ [\mathscr{A}_{i,\alpha}(q) A_i(q) - \mathscr{B}_\alpha B]^2 \cdot \frac{\partial n_i(q)}{\partial \varepsilon_i(q)} - [\mathscr{A}_{i,\alpha}(q) B - \mathscr{B}_\alpha A_i(q)]^2 \cdot \frac{n_i(q)}{\varepsilon_i(q)} \right\} \quad (20)$$

where $\varepsilon_i(q) = [(A_i(q) + B)(A_i(q) - B)]^{\frac{1}{2}}$ and $i = 1$ or 2 corresponds to acoustic and optical modes respectively (see refs. 8 and 9; it should be noted that the exchange integral defined here is one half of that in ref. 9).

For propagation in high symmetry directions, the parameters of equation (20) have the following values:

(I) Longitudinal sound waves in symmetry directions ($\alpha = 1$, 2 and 3 here represents the direction of the k-vector):

$$\mathscr{A}_{i,\alpha}(q) = -2J(J_0 + J_0' - J_q + (-1)^i |J_q'|) + \mathscr{A}_\alpha \quad (21)$$

$$\mathscr{A}_1 = D_{01} + 3D_{22}\cos 2\varphi - 10D_{44}\cos 4\varphi$$
$$\mathscr{B}_1 = D_{01} - D_{22}\cos 2\varphi + 6D_{44}\cos 4\varphi \quad (22)$$

$$\mathscr{A}_2 = D_{01} - 3D_{22}\cos 2\varphi + 10D_{44}\cos 4\varphi$$
$$\mathscr{B}_2 = D_{01} + D_{22}\cos 2\varphi - 6D_{44}\cos 4\varphi \quad (23)$$

$$\mathscr{A}_3 = D_{03}; \quad \mathscr{B}_3 = D_{03} \quad (24)$$

$$\Gamma_1 = \Gamma_2 = D_{22}\sin 2\varphi - 2D_{44}\sin 4\varphi; \Gamma_3 = 0 \quad (25)$$

(II) Transverse sound waves in the basal plane (either in a 1. or 2. direction) with the polarization vector in the basal plane ($\alpha = 6$):

$$\mathscr{A}_{i,6}(q) = \mathscr{A}_6 = 3D_{22}\sin 2\varphi + 10D_{44}\sin 4\varphi;$$
$$\mathscr{B}_6 = -D_{22}\sin 2\varphi - 6D_{44}\sin 4\varphi \quad (26)$$

$$\Gamma_6 = D_{22}\cos 2\varphi + 2D_{44}\cos 4\varphi \quad (27)$$

(III) Transverse sound waves in the c-direction (φ can here be considered as the angle between the polarization vector and the z-axis). This velocity (and change in velocity) is equal to the velocity (change) of the transverse sound waves in the basal plane with the polarization vector parallel to the c-axis:

$$\frac{\Delta v_5}{v_5} = -\frac{H_0^2 c_{44}\cos^2\varphi}{2NJ(A_1(0) + B)} - \frac{H_0^2 c_{44}\sin^2\varphi}{2N^2 J(J - \frac{1}{2})} \sum_{q,i} \frac{n_i(q)}{\varepsilon_i(q)} \quad (28)$$

An external magnetic field (H) will, besides a possible change of the angle φ, modify the strength of the magneto-elastic interaction. This is mainly due to the H-dependence of $\varepsilon_i(q)$:

$$A_i(q, H) = A_i(q, 0) + g\mu_B H \quad (29)$$

which implies:

$$\partial(\Delta v/v)/\partial H = g\mu_B \sum_{q,i} \mathrm{d}(\Delta v/v)/\mathrm{d}A_i(q) \quad (30)$$

so that

$$\alpha_\parallel = [\partial(\Delta v_5/v_5)/\partial H]_{\varphi=0} = H_0^2 c_{44} g\mu_B / [2NJ(A_1(0) + B)^2] \quad (31)$$

and

$$\alpha_\perp = [\partial(\Delta v_5/v_5)/\partial H]_{\varphi=\pi/2} = \frac{H_0^2 c_{44} g\mu_B}{2N^2 J(J - \frac{1}{2})} \sum_{q,i} \frac{A_i(q) n_i(q)}{\varepsilon_i(q)^2} \times \left[\frac{n_i(q) + 1}{k_B T} + \frac{1}{\varepsilon_i(q)} \right] \quad (32)$$

$\Delta = [\Delta v_5/v_5]_{\bar{\varphi}=\pi/2} - [\Delta v_5/v_5]_{\bar{\varphi}=0}$, $\alpha_{\parallel}$ and $\alpha_{\perp}$ have been measured in Terbium at $T = 140°\text{K}$ by Moran and Lüthi,[4] who find:

$$\Delta = 3.0 \,.\, 10^{-3}, \quad \alpha_{\parallel} = 3.6 \,.\, 10^{-5}\ 1/\text{kOe}$$

and

$$\alpha_{\perp} = 7.2 \,.\, 10^{-5}\ 1/\text{kOe}$$

From these experimental values we deduce:

$$H_0 = 1.4 \cdot 10^{-2}, 1/N \sum_{\bar{q}, i} 1/\varepsilon_i(q)^2 \approx 1/[3.3\ \text{meV}]^2$$

and

$$(1/N)\sum_{q, i} 1/\varepsilon_i(q)^3 \approx 1/[3.6\ \text{meV}]^3$$

The values of the energy sums are reasonable, when compared with the experimentally determined dispersion relations for the spin waves.[12,13] The value of H_0 is in fair agreement with the value deduced from magnetostriction measurements[2]: $H_0 = 2.3 \cdot 10^{-2}$ at $T = 140°\text{K}$.

4. THE MAGNON–PHONON INTERACTION

As stated above $\tilde{H}^{(1)}_{S-L}$ gives rise to energy gaps in the magnon energy spectrum. In directions where J'_q is real ($J'_q = J'_{-q}$) interactions only occur between magnons and phonons which are both acoustic or both optical. J'_q is real in the c- and the a- directions, and the Hamiltonian predicts here energy gaps at the nominal crossing points of the magnon and phonon dispersion relations, which have the magnitudes:

$$\Delta\varepsilon_\alpha(\dot{q}) = 2hq\left[\frac{J}{M(A(q) - B)}\right]^{\frac{1}{2}} |\Gamma_\alpha|\,; \quad \alpha = 1 \text{ and } 6 \qquad (33)$$

$$\Delta\varepsilon_5(q) = \hbar q\left[\frac{J}{M(A(q) + B)}\right]^{\frac{1}{2}} \frac{H_0 c_{44}}{NJ} \qquad (34)$$

M is the mass of the atoms. The indices $\alpha = 1, 5$ and 6 refer to the phonon modes defined above. Fig. 1 shows the magnon and the transverse phonon dispersion curves for Terbium in the c-direction at $T = 79°\text{K}$, where $\Delta_1 = 0.6$ meV and $\Delta_2 = 1.5$ meV. Interactions between acoustic longitudinal phonons and acoustic magnons are also observed in the a- and b- directions.[13] Due to the multi-domain character of a ferromagnetic crystal in the absence of a magnetic field, three neutron-groups of equal magnitude should be observed at the crossing points, in a constant $\bar{q}$-scan. One neutron-group arises from domains in which $\varphi = \pi/2$ and $3\pi/2$ (where the magnetization is perpendicular to the q-vector); and two groups show the energy splitting in those domains where $\varphi = \pi/6$, $5\pi/6$, $7\pi/6$ and $11\pi/6$ (see equations (25) and (33)). The neutron data may in fact be satisfactorily interpreted in this manner.[13,14] The expressions (33) and (34) give almost correct

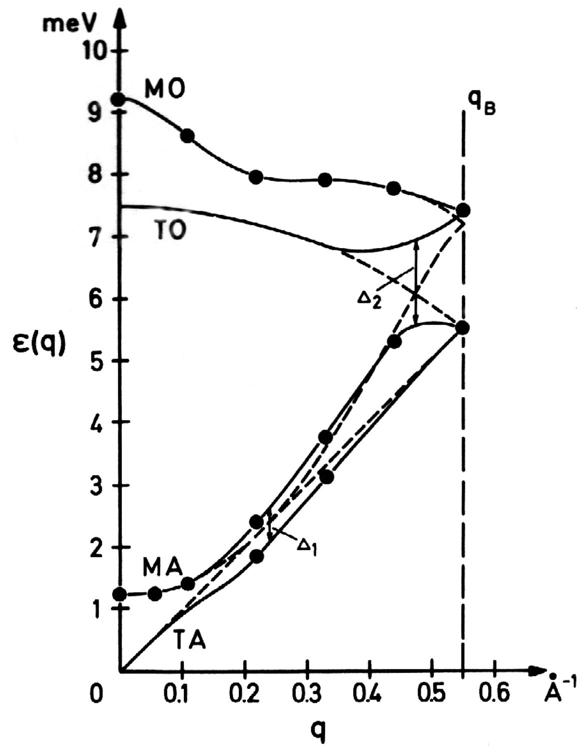

FIGURE 1. The magnon (MA, MO) and transverse phonon (TA, TO) dispersion curves (or Terbium in the c-direction at 79°K. The magnon–phonon interaction causes a mixing of the modes and energy gaps Δ_1 and Δ_2 at the crossing points of the unperturbed dispersion relations (indicated by dashed lines).

values for Δ_1 and the energy splitting in the a-direction. However, the Hamiltonian (1) cannot explain the observation of the energy gap Δ_2 near the Brillouin zone in the c-direction, where an acoustic magnon and an optical phonon dispersion curve cross. The reason why this Hamiltonian is not completely adequate for large wave vectors is probably the use of the concept of "strain" in the one-ion terms. The strain is well defined for small q-vectors, but for large q-vectors the R-differentiation (see (2)) is not uniquely defined. One should use instead the relative displacements of neighbouring ions. This results in no qualitative modifications, but introduces a correction factor, $(2/qc)\sin(qc/2)$, to expression (34), where $\frac{1}{2}c$ is the distance between neighbouring planes in the c-direction (and a similar correction factor in (33). An explanation for the energy splitting Δ_2 requires a direct calculation of the behaviour of the crystal field due to changes in the positions of neighbouring ions.

5. CONCLUSION

Beginning with the magneto-elastic Hamiltonian, we have therefore deduced general expressions for the modification of the velocity of acoustic waves in a ferromagnetic metal due to magnetic ordering and the application of a magnetic field. It has been shown that these expressions give semi-quantitative agreement with the experimental results for Terbium, and they should also be readily applicable to measurements on the other ferromagnetic heavy rare earth metals. The same Hamiltonian, with macroscopic magneto-elastic parameters, also gives an adequate account of the magnon–phonon interactions at short wavelengths which have been observed by inelastic neutron scattering, provided that the interaction is between acoustic phonons and acoustic magnons, or optical phonons and optical magnons. The treatment of other cases requires an extension of the Hamiltonian to include explicitly the relative displacements of neighbouring ions.

We are attempting to reformulate the problem in order to take account of such effects and we also plan to carry out numerical calculations of the magnon sums involved in the expressions for the acoustic velocities, to allow a more detailed comparison with experiments. Further experimental measurements are also being made of the effect of magnetization and field on acoustic velocities in Terbium. The magnon–phonon interaction is being further examined by inelastic neutron scattering measurements in a Terbium crystal which has been made into a single domain by the application of a field. The results of these studies will be reported in due course.

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