Rotational invariance and dipolar magnetoelastic effects: Application to TmSb

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In a magnetic material, the conservation of the total angular momentum is known to give rise to interactions between the magnetic moments and the phonons. The most spectacular effect is the lifting of the degeneracy between transverse sound waves when the directions of propagation and the polarization vectors are interchanged. Here the theory of rotational invariance is reconsidered. It is found that long-wavelength effects appear only in the presence of an external magnetic field. The second-order strain interactions, considered previously in conjunction with the rotational interactions, are found to be eliminated by the equilibrium conditions. The contributions due to finite deformations of the crystal, however, are significant. The magnetic dipole coupling changes rapidly at long wavelengths and it affects the field dependence of the transverse-sound velocities, in a way similar to the rotational interaction terms. The present theory, in which this dipolar contribution is included, gives a much improved account of the ultrasonic experiments performed by Wang and Lüthi on TmSb.

I. INTRODUCTION

The conservation of the total angular momentum of the combined system of localized electronic angular momenta of the ions in a solid, and the angular momenta associated with the rotational part of the vibrations in the crystal, is of fundamental importance. This problem has been discussed several times in the literature. Continuum theories were developed by Tiersten[1] and by Brown.[2] The first microscopic Hamiltonian theory of Melcher[3] was followed by a number of papers[4–9] discussing both various modifications of the theory and experimental findings. In the last decade the subject has not given rise to much discussion. The rotational effects are not very pronounced, and their proper identification requires a considerable effort. Wang and Lüthi[9] did, however, succeed in performing ultrasonic experiments on TmSb, which confirmed, at least qualitatively, the predictions of Dohm and Fulde.[5]

This matter seemed to be settled by the paper of Wang and Lüthi, but a reconsideration of the theory shows that it is unnecessarily complicated and that it still contains some erroneous results. The rotational effects on the field dependence of the transverse sound waves in TmSb, in the case of two different configurations, were predicted to be a factor of 2–3 smaller than the experiments indicated. These large numerical discrepancies are found here to be due to the magnetic dipole interaction, in the one case, and to the variation in the path of the sound waves, caused by relatively large magnetostrictive deformations, in the other case.

In Sec. II the small-strain magnetoelastic theory, which neglects rotational effects, is recapitulated and is extended to account for the presence of the long-range dipole-dipole interaction. In Sec. III the finite-strain contributions are introduced together with the effects derived from the requirement of rotational invariance. The theory is mainly focused on the case of a cubic paramagnet, corresponding to TmSb. In Sec. IV the theoretical predictions are compared with the experimental observations made on TmSb, and the conclusions are given in Sec. V.

II. MAGNETOELASTIC EFFECTS IN RARE-EARTH SYSTEMS IN THE SMALL-STRAIN LIMIT

In this presentation of the influence of rotational invariance on the magnetomechanical properties of a solid we assume a number of simplifying conditions. The magnetic moments of the ions are well localized and are equal to $g\mu_B\mathbf{J}_i$, where $\mathbf{J}_i$ is the angular momentum of the ith ion. In the crystal the ions are subjected to a crystalline electric field of cubic point symmetry, and the two-ion interactions are assumed to be linear in $\mathbf{J}_i$, but are allowed to be anisotropic. Hence, the Hamiltonian for the angular momentum system is assumed to be

$$\mathcal{H}_1(\mathbf{J})=\sum_i\{B_4[O_4^0(\mathbf{J}_i)+5O_4^4(\mathbf{J}_i)]+B_6[O_6^0(\mathbf{J}_i)-21O_6^4(\mathbf{J}_i)]-g\mu_B\mathbf{H}\cdot\mathbf{J}_i\}-\tfrac{1}{2}\sum_{\alpha,\beta}\sum_{i,j}\mathcal{J}_{\alpha\beta}(i,j)J_{i\alpha}J_{j\beta} \quad (1)$$

including the Zeeman term due to the magnetic field $\mathbf{H}$. $O_l^m(\mathbf{J})$ are the Stevens operator equivalents. m positive or negative indicates the operators proportional to $(J_+^{|m|}+J_-^{|m|})/2$ or $(J_+^{|m|}-J_-^{|m|})/2i$, respectively. The x, y, and z axes or, respectively, the 1, 2, and 3 axes are assumed to be along the three cubic directions.

To begin with we neglect the possibility that the crystal might rotate. Hence, we first outline the conventional small-strain theory and then, in the next section, consider the necessary modifications. In the presence of only long-wavelength deformations, the elastic part of the Hamiltonian is

$$\mathcal{H}_2(\overleftrightarrow{\mathbf{E}})=\tfrac{1}{2}\sum_i\{c_\alpha E_\alpha^2(i)+c_\gamma[E_{\gamma1}^2(i)+\tfrac{3}{4}E_{\gamma2}^2(i)] +c_\varepsilon[E_{12}^2(i)+E_{13}^2(i)+E_{23}^2(i)]+\mathbf{p}_i^2/M\}\ , \tag{2}$$

with the inclusion of the kinetic energy of the ions of mass M. Here we have introduced the reduced elastic constants,

$$\begin{aligned} c_\alpha&=\tfrac{1}{3}(c_{11}+2c_{12})V/N\ ,\\ c_\gamma&=4c_{66}V/N=2(c_{11}-c_{12})V/N\ ,\\ c_\varepsilon&=4c_{44}V/N\ , \end{aligned} \tag{3}$$

where N is the number of (magnetic) ions and V the volume. With no stress acting on the system, the mean values of the strains are zero, in which case $E_{kl}(i)$ is equal to the strain tensor at the site i at the time t, $\varepsilon_{kl}=\varepsilon_{kl}(i,t)$, with the short-hand notations,

$$\begin{aligned} \varepsilon_\alpha&=\varepsilon_{11}+\varepsilon_{22}+\varepsilon_{33}\ ,\\ \varepsilon_{\gamma1}&=\tfrac{1}{2}(\varepsilon_{11}-\varepsilon_{22})\ ,\\ \varepsilon_{\gamma2}&=\tfrac{2}{3}\varepsilon_{33}-\tfrac{1}{3}\varepsilon_{11}-\tfrac{1}{3}\varepsilon_{22}\ . \end{aligned} \tag{4}$$

If $\mathbf{r}=\mathbf{r}(i,t)$ is the position of the ith ion and $\mathbf{R}=\mathbf{R}(i)$ the equilibrium position $\mathbf{R}=\langle\mathbf{r}\rangle$, then the displacements are

$$\mathbf{u}(i,t)=\mathbf{r}(i,t)-\mathbf{R}(i) \tag{5a}$$

and the strain tensor is

$$\varepsilon_{kl}=\tfrac{1}{2}(\partial u_k/\partial R_l+\partial u_l/\partial R_k)\ . \tag{5b}$$

The crystalline field is modified if the crystal is strained, and to leading order the crystal-field Hamiltonian is modified linearly in the local deformations. Utilizing the cubic point symmetry, the extra contributions can be written as

$$\mathcal{H}_3(\mathbf{J},\overleftrightarrow{\mathbf{E}})=\sum_i\{G_2[O_2^2(\mathbf{J}_i)E_{\gamma1}(i)+\tfrac{1}{2}O_2^0(\mathbf{J}_i)E_{\gamma2}(i)] +G_3[O_2^{-2}(\mathbf{J}_i)E_{12}(i)+2O_2^1(\mathbf{J}_i)E_{13}(i) +2O_2^{-1}(\mathbf{J}_i)E_{23}(i)]\}\ , \tag{6}$$

including only the lowest, second-rank contributions.

The total Hamiltonian $\mathcal{H}=\mathcal{H}_1(\mathbf{J})+\mathcal{H}_2(\overleftrightarrow{\mathbf{E}})+\mathcal{H}_3(\mathbf{J},\overleftrightarrow{\mathbf{E}})$, which describes the static and dynamic properties in the long-wavelength limit, is first considered in the molecular-field (MF) approximation ($J_{i\alpha}J_{j\beta}$ is replaced by $J_{i\alpha}\langle J_{j\beta}\rangle+\langle J_{i\alpha}\rangle J_{j\beta}-\langle J_{i\alpha}\rangle\langle J_{j\beta}\rangle$), and we assume that all the expectation values are independent of the site considered (para- or ferromagnet with $\langle J_{i\alpha}\rangle=\langle J_\alpha\rangle$). The homogeneous strains are classical quantities, and they are determined by the equilibrium conditions $\langle\partial\mathcal{H}/\partial E_{kl}\rangle=0$ or

$$\begin{aligned} \langle E_{\gamma1}\rangle&=-\frac{1}{c_\gamma}G_2\langle O_2^2(J)\rangle\ ,\\ \langle E_{\gamma2}\rangle&=-\tfrac{2}{3}\frac{1}{c_\gamma}G_2\langle O_2^0(J)\rangle\ ,\\ \langle E_{12}\rangle&=-\frac{1}{c_\varepsilon}G_3\langle O_2^{-2}(J)\rangle\ , \end{aligned} \tag{7}$$

and similar equations for $\langle E_{13}\rangle$ and $\langle E_{23}\rangle$ ($\langle\ \rangle$ are the self-consistent thermal-average values at the temperature T). Notice that in the present approximation $\langle E_\alpha\rangle=0$, because E_α does not appear in $\mathcal{H}_3(\mathbf{J},\overleftrightarrow{\mathbf{E}})$.

Introducing the equilibrium conditions in the total Hamiltonian $\mathcal{H}$ and assuming $E_{kl}=\langle E_{kl}\rangle+\varepsilon_{kl}$, as usual in a small-strain theory, we get

$$\mathcal{H}=\mathcal{H}_1(\mathbf{J})+\mathcal{H}_2(\langle\overleftrightarrow{\mathbf{E}}\rangle)+\mathcal{H}_2(\overleftrightarrow{\boldsymbol{\varepsilon}}) +\mathcal{H}_3(\mathbf{J},\langle\overleftrightarrow{\mathbf{E}}\rangle)+\mathcal{H}_3(\mathbf{J}-\langle\mathbf{J}\rangle,\overleftrightarrow{\boldsymbol{\varepsilon}})\ , \tag{8}$$

where the argument $\mathbf{J}-\langle\mathbf{J}\rangle$ in the last term indicates that $O_2^2(\mathbf{J}_i)$ in (6) should be replaced by $O_2^2(\mathbf{J}_i)-\langle O_2^2\rangle$, and, similarly, for the other terms. The expectation value of $\mathcal{H}_3(\mathbf{J},\langle\overleftrightarrow{\mathbf{E}}\rangle)$ is equal to $-2\mathcal{H}_2(\langle\overleftrightarrow{\mathbf{E}}\rangle)$ and this term describes the contributions to the magnetic part of the Hamiltonian which are proportional to the equilibrium strains. The MF part of $\mathcal{H}_1(\mathbf{J})+\mathcal{H}_3(\mathbf{J},\langle\overleftrightarrow{\mathbf{E}}\rangle)$ is diagonalized, subject to conditions (7), determining the $(2J+1)$ eigenvalues $\mathcal{E}_\nu$ and the corresponding eigenstates $|\nu\rangle$. The two-ion part of $\mathcal{H}_1(\mathbf{J})$ is expanded in terms of the excitation operators of this basis, $a_{\nu\mu}(i)=(|\nu\rangle\langle\mu|)_i$. Without the last term in (8), the equations of motion for the Green functions constructed from these operators are diagonalized straightforwardly, utilizing a random-phase decoupling. This procedure leads to the usual RPA result for the generalized susceptibility. The nonequilibrium coupling term $\mathcal{H}_3(\mathbf{J}-\langle\mathbf{J}\rangle,\overleftrightarrow{\boldsymbol{\varepsilon}})$ is linear in both the operators $a_{\nu\mu}(i)$ and the phonon operators $\beta_{\mathbf{q},s}$ introduced via (5b) and

$$\mathbf{u}(i)=\sum_{\mathbf{q},s}\mathbf{F}(\mathbf{q},s)(\beta_{\mathbf{q},s}+\beta^\dagger_{-\mathbf{q},s})e^{i\mathbf{q}\cdot\mathbf{R}(i)} \tag{9a}$$

with

$$\mathbf{F}(\mathbf{q},s)=\hbar^{1/2}[2NM\omega_s(\mathbf{q})]^{-1/2}\mathbf{f}(\mathbf{q},s)\ . \tag{9b}$$

$\omega_s(\mathbf{q})$ is the eigenfrequency of the sth phonon mode at the wave vector $\mathbf{q}$, and $\mathbf{f}(\mathbf{q},s)$ is the polarization unit vector of this mode. The coupled equations of motion for the magnetic and the phonon Green functions are deduced, and are linearized as above by the random-phase decoupling and a Fourier transformation. The energies of the normal modes are then the roots of the determinant of the linear set of equations or, equivalently, the poles of the Green functions.

The amplitude of the coupling between the magnetic excitations and the phonons goes to zero (as $q[\omega_s(\mathbf{q})]^{-1/2}$) when q goes to zero, hence the energies of the uniform modes are not changed. However, the slopes of the low-frequency dispersion relations of the nearly pure phononlike excitations are modified, corresponding

to a change of the elastic constant $M(N/V)[\omega_s(\mathbf{q})/q]^2$. Considering for instance the transverse sound waves propagating along one of the cube axes, the result is

$$\Delta c_{44}/c_{44} = -\frac{1}{c_\varepsilon}\Xi_{\mathbf{q}}(G_3 O_2^{-2}) , \tag{10}$$

$\Xi_{\mathbf{q}}(A)$ is a zero-frequency susceptibility, which may, however, depend on the wave vector of the sound wave considered. The zero-frequency susceptibility of the operator set (A,B) is determined from the MF Hamiltonian as

$$\chi(A,B) = \sum_{\substack{\nu,\mu \\ (\mathcal{E}_\nu \neq \mathcal{E}_\mu)}} \langle \nu | A | \mu \rangle \langle \mu | B | \nu \rangle \frac{n_\nu - n_\mu}{\mathcal{E}_\mu - \mathcal{E}_\nu} + \frac{1}{k_B T}\left[\sum_{\substack{\nu,\mu \\ (\mathcal{E}_\nu = \mathcal{E}_\mu)}} \langle \nu | A | \mu \rangle \langle \mu | B | \nu \rangle n_\nu - \langle A \rangle \langle B \rangle \right] , \tag{11}$$

where n_ν is the population of the νth MF level, in terms of which

$$\Xi_{\mathbf{q}}(A) = \chi(A,A) + \sum_\alpha \chi^2(A,J_\alpha)\mathcal{J}_{\alpha\alpha}(\mathbf{q})\{1-\chi(J_\alpha,J_\alpha)\mathcal{J}_{\alpha\alpha}(\mathbf{q})\}^{-1} , \tag{12}$$

assuming the Fourier transform of $\mathcal{J}_{\alpha\beta}(ij)$ and $\chi(J_\alpha,J_\beta)$ to be diagonal with respect to the (xyz) axes. Equations (10)–(12) constitute the result also obtained in Ref. 10 (see also Thalmeier and Fulde[11]) and they reduce to the result of, for instance, Dohm and Fulde[5] when $\mathcal{J}_{\alpha\alpha}(\mathbf{q}\approx 0)$ can be neglected. Finite frequency corrections are negligible as long as $\hbar^2\omega_s^2(\mathbf{q}) \ll (\mathcal{E}_\nu - \mathcal{E}_\mu)^2$, $\mathcal{E}_\nu \neq \mathcal{E}_\mu$, which condition may also effectively determine the regime where the last elastic term in (11) stays constant.

The $\mathbf{q}$ dependence of $\mathcal{J}_{\alpha\beta}(\mathbf{q})$ in (12) would normally be entirely negligible at the small wave vectors employed in, for instance, an ultrasonic experiment. This statement is valid for all couplings except the magnetic dipole-dipole interaction, which changes extremely rapidly around zero wave vector. Although it is rather weak, the abrupt changes of this coupling near $\mathbf{q}=0$ lead to jumps in the magnetic excitation spectrum which have recently been observed in $Ho_{90}Tb_{10}$ in both its ferromagnetic and helical magnetic phases.[12] In the present context the two-ion coupling is considered to comprise an isotropic Heisenberg exchange interaction and the magnetic dipole coupling, and its Fourier transform is

$$\mathcal{J}_{\alpha\beta}(\mathbf{q}) = \mathcal{J}(\mathbf{q})\delta_{\alpha\beta} + (g\mu_B)^2(N/V)D_{\alpha\beta}(\mathbf{q}) . \tag{13}$$

At long wavelengths $\mathcal{J}(\mathbf{q})$ may be replaced by $\mathcal{J}(\mathbf{0})$, and additional anisotropic two-ion couplings may be considered to be included effectively in the crystal-field Hamiltonian. The dipole coupling is

$$D_{\alpha\beta}(ij) = (V/N)[3R_\alpha(i)R_\beta(j) - R_{ij}^2]/R_{ij}^5 , \tag{14}$$

where $R_{ij} = |\mathbf{R}(i) - \mathbf{R}(j)|$, and in a cubic system[13] at long wavelengths the diagonal Fourier components are

$$D_\parallel(\mathbf{q}) = -8\pi/3; \quad D_\perp(\mathbf{q}) = 4\pi/3 . \tag{15}$$

$D_\parallel$ is the component along the direction of $\mathbf{q}$, and $D_\perp$ gives the two transverse components. (15) is valid as long as $2\pi/L \ll q \ll 2\pi/a$, where L is the length of the crystal and a the lattice parameter (a more detail discussion is given by Keffer in Ref. 13), which is just the regime probed in an ultrasonic experiment. In the paramagnetic case at zero field, $\chi(A,J_\alpha)$ in (12) vanishes due to time-reversal symmetry, A being a quadrupole operator. In the presence of an external field or in an ordered state, $\chi(A,J_\alpha)$ depends strongly on α, which in combination with the anisotropic behavior of $D_{\alpha\beta}(\mathbf{q})$ introduces a directional dependence of $\Xi_{\mathbf{q}}$. In the small-strain limit the dynamic part of the Hamiltonian, (8), only involves the phonon coordinates through $\varepsilon_{kl} = \varepsilon_{lk}$. This means that the velocities of the transverse sound waves for which the directions of the polarization and propagation vectors are interchanged, corresponding to an interchange of k and l, are equal, except for the dependence of $\Xi_{\mathbf{q}}$ on the direction of $\mathbf{q}$ relatively to the direction of the moments. This directional effect associated with the dipole coupling, has previously[3–9] been ascribed to be due to rotational interactions alone. The two mechanisms may produce effects of the same order of magnitude in paramagnetic systems. In the case of a ferromagnet with large magnetic moments, the dipole contribution may strongly dominate as is the case of Terbium, where the effect is clearly observed experimentally and where the mechanism has been properly identified.[14]

III. ROTATIONAL INVARIANCE AND FINITE-STRAIN EFFECTS

In the preceding section the finite-strain contributions to the MF Hamiltonian are included already via $\mathcal{H}_3(\mathbf{J}, \langle \overleftrightarrow{E} \rangle)$ in (8). Here we examine more carefully the expression for $\overleftrightarrow{E}$ when $\langle \overleftrightarrow{E} \rangle$ is nonzero, and we allow for rotational displacements of the crystal. As discussed in the previous papers, see, e.g., Ref. 5, the propagation of a transverse phonon mode implies that both the symmetric and the antisymmetric part of the time-dependent strain tensor $\overleftrightarrow{v}(i,t)$ are nonzero. Introducing

$$\overleftrightarrow{v}(i,t) = \overleftrightarrow{\varepsilon}(i,t) + \overleftrightarrow{\omega}(i,t) , \tag{16a}$$

where the antisymmetric part is

$$\omega_{kl} = \tfrac{1}{2}(\partial u_k/\partial R_l - \partial u_l/\partial R_k) , \tag{16b}$$

the position of the ith ion may be written as

$$\mathbf{r}(i,t) = \mathbf{R}(i) + \mathbf{u}(i,t) = [\overleftrightarrow{1} + \overleftrightarrow{v}(i,t)]\cdot\mathbf{R}(i) \tag{17}$$

in the linear regime, neglecting the second derivatives of $\mathbf{u}$ ($q \ll 2\pi/a$). For simplification we restrict ourselves to

the case where the displacements $\mathbf{u}(i,t)$ all lie in the plane perpendicular to the z axis, corresponding to phonons which have their propagation and polarization vectors parallel to this plane. Hence we assume

$$\overleftrightarrow{\mathbf{v}}(i,t)=\begin{bmatrix} \varepsilon_{11} & \varepsilon_{12}+\omega_{12} & 0 \\ \varepsilon_{12}-\omega_{12} & \varepsilon_{22} & 0 \\ 0 & 0 & 0 \end{bmatrix} . \tag{18}$$

A constant ω_{12} describes a rotation of the crystal around the z axis by an angle $-\theta$, where $\tan\theta=\omega_{12}[1+(\varepsilon_{11}+\varepsilon_{22})/2]^{-1}$ or $\theta=\omega_{12}$ to leading order.

The Hamiltonians (2) and (6) are established by general symmetry arguments referring to the unstrained lattice, and the deformation matrix $\overleftrightarrow{\mathbf{E}}$ is determined with reference to the atomic positions $\mathbf{R}^0(i)$ of the unstrained lattice, not to the equilibrium positions

$$\mathbf{R}(i)=(\overleftrightarrow{1}+\langle\overleftrightarrow{\mathbf{E}}\rangle)\cdot\mathbf{R}^0(i) . \tag{19}$$

Furthermore, it cannot include terms which correspond to a rigid rotation of the crystal, which does not involve any change in the distances between the ions. According to this we shall write

$$\mathbf{r}(i,t)=\overleftrightarrow{\mathcal{R}}(\theta)\cdot(\overleftrightarrow{1}+\overleftrightarrow{\mathbf{E}})\cdot\mathbf{R}^0(i) , \tag{20}$$

where $\overleftrightarrow{\mathcal{R}}(\theta)$ is the transformation matrix rotating a vector (at site i) through the angle $-\theta$ around the z axis:

$$\overleftrightarrow{\mathcal{R}}(\theta)=\begin{bmatrix} \cos\theta & \sin\theta & 0 \\ -\sin\theta & \cos\theta & 0 \\ 0 & 0 & 0 \end{bmatrix} . \tag{21}$$

Given $\overleftrightarrow{\mathbf{v}}(i,t)$ the easiest way to proceed is to assume that $\overleftrightarrow{\mathbf{E}}$ in (20) is a symmetric matrix. This assumption is justified through the general finite-strain theory, see Refs. 2, 3, 5 and references therein. The symmetric deformation matrix $\overleftrightarrow{\eta}$, defined by

$$\eta_{kl}=\frac{1}{2}\left[\frac{\partial u_k^0}{\partial R_l^0}+\frac{\partial u_l^0}{\partial R_k^0}+\sum_m \frac{\partial u_m^0}{\partial R_k^0}\frac{\partial u_m^0}{\partial R_l^0}\right] , \tag{22a}$$

where $\mathbf{u}^0=\mathbf{r}-\mathbf{R}^0$, can be shown to be invariant under a rigid rotation of the lattice, and

$$\mathbf{r}=\overleftrightarrow{\mathcal{R}}(\theta)\cdot(\overleftrightarrow{1}+2\overleftrightarrow{\eta})^{1/2}\cdot\mathbf{R}^0(i) , \tag{22b}$$

hence $\overleftrightarrow{\mathbf{E}}=\overleftrightarrow{\eta}-\frac{1}{2}\overleftrightarrow{\eta}^2+\cdots$ is a symmetric matrix. Combining Eqs. (17)–(20) we have

$$\overleftrightarrow{1}+\overleftrightarrow{\mathbf{E}}=\overleftrightarrow{\mathcal{R}}^{-1}(\theta)\cdot(\overleftrightarrow{1}+\overleftrightarrow{v})\cdot(\overleftrightarrow{1}+\langle\overleftrightarrow{\mathbf{E}}\rangle) \tag{23}$$

with θ determined so that $\overleftrightarrow{\mathbf{E}}$ is symmetric, implying

$$\theta=\omega_{12}+\tfrac{1}{2}\sum_m(\varepsilon_{1m}\langle E_{m2}\rangle-\varepsilon_{2m}\langle E_{m1}\rangle) \tag{24}$$

to leading order in the small strains $\overleftrightarrow{\mathbf{v}}$, neglecting terms of second order in the equilibrium strains. Within the same accuracy

$$E_{kl}-\langle E_{kl}\rangle=\varepsilon_{kl}+\tfrac{1}{2}\sum_m(\varepsilon_{km}\langle E_{ml}\rangle+\varepsilon_{lm}\langle E_{mk}\rangle) . \tag{25}$$

The Hamiltonians (2) and (6) are derived from an expansion in powers of $\overleftrightarrow{\mathbf{E}}$, and in order to restore the validity of this expansion, when θ is nonzero, the vectors in the system $\mathbf{r}$, $\mathbf{J}$, and $\mathbf{H}$ (and $\mathbf{p}$) are replaced by $\overleftrightarrow{\mathcal{R}}^{-1}(\theta)\cdot\mathbf{r}$, $\overleftrightarrow{\mathcal{R}}^{-1}(\theta)\cdot\mathbf{J}$, and $\overleftrightarrow{\mathcal{R}}^{-1}(\theta)\cdot\mathbf{H}$ [and $\overleftrightarrow{\mathcal{R}}^{-1}(\theta)\cdot\mathbf{p}$], respectively. This transformation is valid on account of conservation of the total angular momentum for the whole system under consideration (including the sources of the external fields). Another way to express this is that the requirement that the whole system should be rotationally invariant implies that the Hamiltonian considered in Sec. II can only be valid when $\theta=0$. As soon as θ is nonzero new terms have to appear, which are obtained by replacing $O_l^m(\mathbf{J})$ by

$$\begin{aligned} O_l^m(\overleftrightarrow{\mathcal{R}}^{-1}(\theta)\cdot\mathbf{J})&=e^{i\theta J_z}O_l^m(\mathbf{J})e^{-i\theta J_z} \\ &=O_l^m(\mathbf{J})\cos(m\theta)-O_l^{-m}(\mathbf{J})\sin(m\theta) \end{aligned} \tag{26}$$

in all terms in $\mathcal{H}$, and also replacing $\mathbf{H}$ by $\overleftrightarrow{\mathcal{R}}^{-1}(\theta)\cdot\mathbf{H}$, implying that the Zeeman term is unchanged (the same is the case with the kinetic-energy term). The argumentation is strictly valid only if θ is constant throughout the crystal. However, it remains valid in the inhomogeneous case, with θ replaced by its local value θ_i, as long as the modulation wavelength is long compared to the range of the microscopic interactions (i.e., $q\ll 2\pi/a$). With the use of (26) the extra contributions to the total Hamiltonian introduced when θ_i is nonzero:

$$\begin{aligned} \Delta\mathcal{H}(\theta)=&\mathcal{H}_1(\overleftrightarrow{\mathcal{R}}^{-1}(\theta)\cdot\mathbf{J},\overleftrightarrow{\mathcal{R}}^{-1}(\theta)\cdot\mathbf{H})-\mathcal{H}_1(\mathbf{J},\mathbf{H}) \\ &+\mathcal{H}_3(\overleftrightarrow{\mathcal{R}}^{-1}(\theta)\cdot\mathbf{J},\overleftrightarrow{\mathbf{E}})-\mathcal{H}_3(\mathbf{J},\overleftrightarrow{\mathbf{E}}) \end{aligned} \tag{27}$$

can be written down without difficulty. The equilibrium conditions have to be supplemented with $\langle\partial\mathcal{H}/\partial\theta\rangle=0$. The implicit assumption that $\langle\theta\rangle=0$, as made in (23) or (24), can be considered to be valid in general. $\langle\theta\rangle$ being nonzero means that the external field is applied in a different direction than intended, and one merely has to provide a mechanical torque in order to assure that $\langle\theta\rangle=0$. With $\langle\theta\rangle=0$ the equilibrium Eqs. (7) are unchanged. The modifications are then purely dynamic, and they amount to replacing $\overleftrightarrow{\varepsilon}$ in $\mathcal{H}_2(\overleftrightarrow{\varepsilon})$ and $\mathcal{H}_3(J-\langle J\rangle,\overleftrightarrow{\varepsilon})$ in the total Hamiltonian (8) by $\overleftrightarrow{\mathbf{E}}-\langle\overleftrightarrow{\mathbf{E}}\rangle$ given by (25), and adding $\Delta\mathcal{H}(\theta)$ (27), to this Hamiltonian.

$\Delta\mathcal{H}(\theta)$ is expanded to second order in θ_i, with θ given by (24). As first realized by Dohm and Fulde[5] the terms of second order contribute to the phonon energies in the same order of magnitude as the first-order terms. In the situation where $\mathbf{H}$ is applied along one of the cubic axes only the strains $\langle E_{\gamma 1}\rangle$ and $\langle E_{\gamma 2}\rangle=\langle E_{33}\rangle=-(\langle E_{11}\rangle+\langle E_{22}\rangle)$ are different from zero. Further, if only transverse phonons with $\mathbf{q}$ parallel to the 1 axis and $\mathbf{f}$ parallel to the 2 axis, or with $\mathbf{q}$ and $\mathbf{f}$ interchanged, are excited, then the only nonzero dynamic strain parameters are ε_{12} and ω_{12}, where $\varepsilon_{12}=-\omega_{12}$ or $\varepsilon_{12}=\omega_{12}$, respectively. The relevant phonon part of the Hamiltonian, linear and quadratic in ε_{12} and ω_{12}, describing this situation is to leading order in the equilibrium strains:

$$\sum_i \{\tfrac{1}{2}c_\varepsilon \varepsilon_{12}^2(i)(1-\langle E_{\gamma 2}\rangle)+G_3' O_2^{-2}(\mathbf{J}_i)\varepsilon_{12}(i) -[\Omega_1(i)-2G_3'' O_2^2(\mathbf{J}_i)\varepsilon_{12}(i)][\omega_{12}(i)-\langle E_{\gamma 1}\rangle\varepsilon_{12}(i)]-\tfrac{1}{2}\Omega_2(i)[\omega_{12}^2(i)-2\langle E_{\gamma 1}\rangle\varepsilon_{12}(i)\omega_{12}(i)]\} \quad (28)$$

when introducing the two operators

$$\Omega_1=20B_4O_4^{-4}-84B_6O_6^{-4}+2G_2O_2^{-2}\langle E_{\gamma 1}\rangle-2G_3O_2^2\langle E_{12}\rangle \quad (29a)$$

and

$$\Omega_2=80B_4O_4^4-336B_6O_6^4+4G_2O_2^2\langle E_{\gamma 1}\rangle+4G_3O_2^{-2}\langle E_{12}\rangle \quad (29b)$$

and defining

$$G_3'=G_3(1-\tfrac{1}{2}\langle E_{\gamma 2}\rangle);\quad G_3''=G_3' \ . \quad (29c)$$

In (28) we have neglected the contribution of the two-ion interaction to $\Delta\mathcal{H}(\theta)$. This is proportional to $\mathcal{J}_{\alpha\beta}(\mathbf{q})-\mathcal{J}_{\alpha\beta}(\mathbf{0})$, so that only the magnetic dipole coupling contributes and this is a minute term. According to the procedure of Sec. II the Hamiltonian above leads to the following change of the "dynamic" elastic constant:

$$\Delta c_{44}/c_{44}=-\frac{1}{c_\varepsilon}\{\Xi_{\mathbf{q}}[G_3'O_2^{-2}\pm\Omega_1(1\pm\langle E_{\gamma 1}\rangle)]+\langle\Omega_2\rangle(1\pm2\langle E_{\gamma 1}\rangle)\pm4G_3''\langle O_2^2\rangle(1\pm\langle E_{\gamma 1}\rangle)\}-\langle E_{\gamma 2}\rangle \ , \quad (30)$$

where the upper signs (+) refer to the case where $\mathbf{q}\|1$ axis and the lower ones to $\mathbf{q}\|2$ axis. Considering instead the situation where $\mathbf{H}\|(1,1,0)$ and where $\mathbf{q}$ and $\mathbf{f}$ are parallel to, respectively, (1,1,0) and (1,−1,0), then we get the following result with the upper signs:

$$\Delta c_{66}/c_{66}=-\frac{1}{c_\gamma}\{\Xi_{\mathbf{q}}[-G_2'O_2^2\pm\Omega_1(1\pm\langle E_{12}\rangle)]+\langle\Omega_2\rangle(1\pm2\langle E_{12}\rangle)\pm4G_2''\langle O_2^{-2}\rangle(1\pm\langle E_{12}\rangle)\}-\langle E_{\gamma 2}\rangle \ , \quad (31)$$

whereas the lower signs (−) apply to the case where the directions of $\mathbf{q}$ and $\mathbf{f}$ are interchanged. The primes on G_2 have the same meanings as in (29c), $G_2'=G_2(1-\frac{1}{2}\langle E_{\gamma 2}\rangle)$; $G_2''=G_2'$. Neglecting the contributions of the dipole coupling and the terms proportional to the equilibrium strains (and B_6), these two results reduce to those derived by Dohm and Fulde.[5] They (and Melcher and coworkers[7,8]) also include contributions arising from the terms linear in $\vec{\mathbf{E}}-\langle\vec{\mathbf{E}}\rangle$ but of second-order in the dynamic strain variables (appearing when $\vec{\mathbf{E}}$ is replaced by $\vec{\eta}$). However, these contributions are simply cancelled by the equilibrium conditions. The expectation value of the prefactor to $\vec{\mathbf{E}}-\langle\vec{\mathbf{E}}\rangle$ in the Hamiltonian vanishes at equilibrium, as does the expectation value of the prefactor to ω_{12} in (28) ($\langle\Omega_1\rangle=0$ or $\langle\theta\rangle=0$ is not properly fulfilled). In these papers[5–9] the dynamic Hamiltonian linear in the deformations is considered (effectively) to be $\mathcal{H}_3(\mathbf{J},\vec{\eta}-\langle\vec{\eta}\rangle)$, which should rather be $\mathcal{H}_3(\mathbf{J}-\langle\mathbf{J}\rangle,\vec{\eta}-\langle\vec{\eta}\rangle)$ at equilibrium, corresponding to (8). There is no principal reason for preferring either $\vec{\eta}$ or $\vec{\mathbf{E}}=(1+2\vec{\eta})^{1/2}-1$ as the expansion parameter, as both are invariant to a rigid rotation of the lattice. The results depend on this choice, but it is only the finite-deformation contributions which are affected. $\langle E_{\gamma 2}\rangle$ is replaced by $2\langle E_{\gamma 2}\rangle$ (or rather $2\langle\eta_{\gamma 2}\rangle$) in Eqs. (28)–(31) if we substitute $\vec{\eta}$ for $\vec{\mathbf{E}}$ in the Hamiltonian. This difference just reflects the fact that the parameters occurring to next order in the strains, i.e., the third-order elastic and the second-order magnetoelastic constants, depend on this choice. Our motivation for choosing $\vec{\mathbf{E}}$ is that it is the more familiar quantity in small-strain theories; for instance the third-order elastic constants, are normally defined in terms of these strains. The finite-strain terms in (30) and (31) may be taken as a measure for the modifications which may occur due to the higher-order strain terms in the Hamiltonian. This suggests that only the third-order elastic constants may be of importance. They introduce terms corresponding to the rather trivial contribution $-\langle E_{\gamma 2}\rangle$ appearing both in (30) and (31).

In the paramagnetic phase, at zero field, the terms in (30) and (31) which change sign when the directions of $\mathbf{q}$ and $\mathbf{f}$ are interchanged have to cancel each other by symmetry. Numerical calculations show that this cancellation applies to all terms introduced by $\Delta\mathcal{H}(\theta)$, and that this is a more general feature than indicated by symmetry arguments. For instance, it also occurs in the presence of a tetragonal contribution $BO_2^2(\mathbf{J}_i)$ to $\mathcal{H}_1(\mathbf{J})$ (or, correspondingly, in the presence of an external stress), and in the ferromagnetic case. The reason may be traced back to the manner in which $\Delta\mathcal{H}(\theta)$ was constructed, (26) and (27). Apart from the kinetic and the Zeeman energies, the total Hamiltonian $\mathcal{H}(\theta)=\mathcal{H}(\theta=0)+\Delta\mathcal{H}(\theta)$ can be obtained from $\mathcal{H}(\theta=0)$ by a unitary transformation:

$$\mathcal{H}(\theta)=U\mathcal{H}(\theta=0)U^{-1}+O(\omega^{3/2}) \quad (32a)$$

with

$$U=\exp\left[i\sum_i\theta_iJ_{zi}\right] \quad (32b)$$

as long as $\mathbf{H}=\mathbf{0}$. This means that, in the limit of long wavelengths at low frequencies, where the kinetic corrections proportional to $\omega^{3/2}$ can be neglected, the dependence on θ_i can be removed from $\mathcal{H}(\theta)$ when $\mathbf{H}=\mathbf{0}$, by performing the reciprocal unitary transformation. Therefore the effects of $\Delta\mathcal{H}(\theta)$ on the acoustic sound waves

must vanish at zero magnetic field. A slow rotation of the lattice causes no energy changes as long as there is no external magnetic field to define the amount of rotation (the magnetic system adjusts itself so as to follow the slow rotation adiabatically). The reciprocal transformation may be utilized also when $\mathbf{H}\neq\mathbf{0}$ for deriving alternative expressions for the effects of the rotational interactions. The results are derived straightforwardly and amount to the replacement of the quantities given by (29) in Eqs. (30) and (31) by

$$\begin{aligned}\Omega_1&=g\mu_B(H_1J_2-H_2J_1)\ ,\\ \Omega_2&=-g\mu_B\mathbf{H}\cdot\mathbf{J}\ ,\\ G_2''&=G_3''=0\ ,\end{aligned}\tag{29'}$$

(G_2' and G_3' are unchanged). This alternative result is more transparent and easier to generalize than the original one. In fact it is more correct as it includes, implicitly, the small rotational effects of the dipole coupling neglecting in Eqs. (28) and (29). The introduction of (29') instead of (29) shows that the rotational effects on the sound velocities are quadratic in the magnetic field, at low fields in the paramagnetic phase. In this case the cubic symmetry implies that the rotational interactions contribute exclusively to the velocity difference between the two related transverse sound waves, if the dipole coupling is neglected.

IV. APPLICATION TO TmSb

TmSb is very well suited for studying the effects of rotational invariance. The Tm ions ($J=6$ and $g=\frac{7}{6}$) are positioned on a fcc lattice, with the lattice parameter $a=6.084$ Å, corresponding to $N/V=1.78\times10^{22}$ Tm-ions/cm^3. It is paramagnetic down to the lowest temperatures (singlet ground-state system), and the exchange interaction is found to be negligible. This was concluded from susceptibility measurements[15] and from neutron-scattering experiments,[16] through which the values $B_4=1.12\times10^{-3}$ meV and $B_6=-2.46\times10^{-6}$ meV were determined. The two magnetoelastic parameters G_2 and G_3 have been derived from the temperature variation of c_{44} and c_{66}, relative to that observed in nonmagnetic LaSb by Wang and Lüthi[9] and Mullen *et al.*[17] They found $G_2=-5.43$ meV and $G_3=1.72$ meV. The signs of the two parameters are those indicated by point-charge calculations, and the sign of G_2 has been checked by measuring[18] the magnetostriction along an (100) axis parallel to an applied field of 15 kOe. The magnetostriction $\Delta l/l$ is found to be nearly constant between 2–12 K with a mean value of 1.2×10^{-5}, which is, however, nearly a factor of 2 larger than predicted by the value for G_2 stated above. The estimated demagnetization field is about 1 kOe so that, assuming the internal field to be 14 kOe, the magnetostriction data indicate $G_2=-9.60$ meV. The elastic constant c_{66} exhibits a very rapid variation between 5–20 K, see Fig. 1. In this narrow temperature interval the nonmagnetic c_{66} stays constant, so the rapid change of c_{66} can only be ascribed to crystal-field effects. It turns out that it is reproduced only if the magnitude of G_2 is increased to be about the same as indicated by the magnetostriction measurements. Using $G_2=-9.60$ meV, we obtained the fit shown in Fig. 1 to the temperature variation of $c_{11}-c_{12}=2c_{66}$ measured by Mullen *et al.*[17] We assume that the nonmagnetic value of $c_{11}-c_{12}$ varies smoothly with temperature as shown in the figure. This variation is greater than indicated by the scaling of the LaSb results,[17] also shown in Fig. 1. However, the absolute value of $c_{11}-c_{12}$ is nearly a factor of 2 larger in TmSb than in LaSb, making the scaling assumption applied by Mullen *et al.* somewhat questionable. The low-temperature behavior of $c_{11}-c_{12}$ combined with the magnetostriction data provide a firmer basis for our analysis, and we shall use $G_2=-9.60$ meV in the following. Wang and Lüthi[9] accounted almost perfectly for the temperature dependence of c_{44} using $G_3=1.72$ meV. We have chosen the slightly higher value $G_3=1.85$ meV which still accounts well for c_{44} as a function of T, recalling that the variation of the nonmagnetic value of c_{44} (which is about 30% larger than in LaSb) is somewhat uncertain.

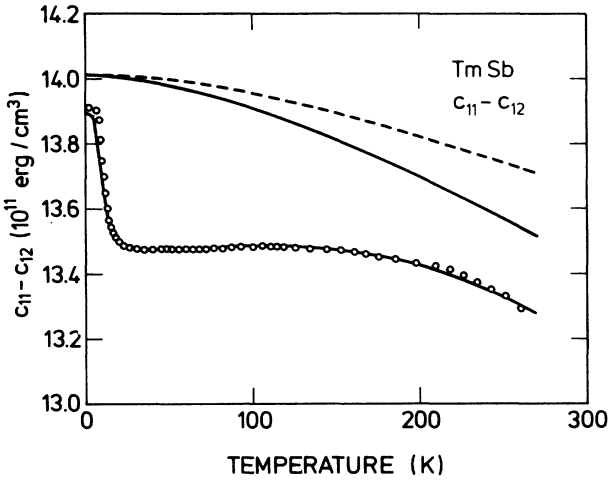

FIG. 1. The elastic constant $c_{11}-c_{12}=2c_{66}$ in TmSb as a function of temperature. The circles are the experimental results of Mullen *et al.* (Ref. 17) and the associated line is the calculated behavior obtained with $G_2=-9.60$ meV. The other solid line shows the assumed variation of the nonmagnetic $c_{11}-c_{12}$. The dashed line is the same quantity obtained from a scaling of the experimental results on LaSb of Mullen *et al.* (Ref. 17).

In order to agree with the analysis of the magnetic susceptibility measurements, including the dipolar coupling, we assume

$$\mathcal{J}_\perp(\mathbf{q}\approx\mathbf{0})=\mathcal{J}(\mathbf{0})+(g\mu_B)^2D_\perp(\mathbf{q}\approx\mathbf{0})N/V=0\ ,$$

as this is the parameter determining the molecular field (when the demagnetization field H_D is subtracted from the applied field H_A). This means that we use

$$\begin{aligned}\mathcal{J}_\parallel(\mathbf{q}\approx\mathbf{0})&=-4\pi(g\mu_B)^2N/V\ ,\\ \mathcal{J}_\perp(\mathbf{q}\approx\mathbf{0})&=0\ ,\end{aligned}\tag{33}$$

where $\mathcal{J}_\parallel(\mathbf{q}\approx\mathbf{0})=-0.0163$ meV in TmSb.

With these values for the parameters we have calculated the field dependences of c_{44} and c_{66} at $T=2$ K. In order to make a comparison with the ultrasonic measurements of Wang and Lüthi we need to introduce two corrections. The first concerns the demagnetization-field correction to the internal field **H** considered in the equations above. The applied field is

$$\mathbf{H}_A=\mathbf{H}+\mathbf{H}_D;\quad \mathbf{H}_D=N_D g\mu_B\langle\mathbf{J}\rangle N/V\ . \tag{34}$$

Wang and Lüthi state that the demagnetization factor N_D is about 4 in their measurements. The correction is small so the precise value is not important, and we use $N_D=4$ in all the calculations, implying that H_D is about 2.5 kOe at maximum field. The other correction is due to magnetostriction, which changes the distances over which the travelling times of the sound waves are measured. The experimental changes of the velocities derived by Wang and Lüthi are not corrected for this effect. They do consider it in the case of the c_{44} modes, where they estimate the length changes to be unimportant. We agree with this conclusion, but when the field is applied along (1,1,0), $\langle E_{12}\rangle\cong -0.6\times10^{-3}$ at 60 kOe leading to important corrections. The nominal velocity change, found by Wang and Lüthi, is determined in terms of the real change by

$$(\Delta v/v)_{\text{nominal}}=\Delta v/v-\Delta l/l\ , \tag{35a}$$

where $\Delta v/v$ is half the relative change of the elastic constant, and

$$\Delta l/l=\begin{cases}\langle E_{12}\rangle-\frac{1}{2}\langle E_{\gamma 2}\rangle \text{ for } \mathbf{q}\|\mathbf{H}\ ,\\ -\langle E_{12}\rangle-\frac{1}{2}\langle E_{\gamma 2}\rangle \text{ for } \mathbf{q}\perp\mathbf{H}\ ,\end{cases} \tag{35b}$$

when $\mathbf{H}\|(1,1,0)$, and the analogous relations when $\mathbf{H}\|(1,0,0)$. With the two corrections included we show in Figs. 2 and 3 the experimental results for $(\Delta v/v)_{\text{nominal}}$ obtained by Wang and Lüthi[9] in comparison with our calculations.

When the field is applied along (1,0,0) the equilibrium strains are small and their contributions to the (nominal) velocity changes in Fig. 2 are insignificant. Instead the rotational interactions, which contribute chiefly to the difference between the two sound velocities, are important. $\mathcal{H}_{\|}(\mathbf{q}\approx\mathbf{0})$ only affects the velocity of the sound waves for which $\mathbf{q}\perp\mathbf{H}$ [because $\chi(A,J_\alpha)=0$ when J_α is the component parallel to **H**]. Hence, the dipole coupling contributes to the splitting between the two velocities, and as indicated in the figure the two mechanisms contribute nearly equally. The agreement found in Fig. 2 is good, with a predicted splitting about 10% larger than observed. For the c_{44} modes, where both the polarization and the propagation vectors are perpendicular to $\mathbf{H}\|(1,0,0)$, the changes are predicted to be about the same as for the c_{44} mode with $\mathbf{q}\perp\mathbf{H}$ in Fig. 2, but of opposite sign. For the c_{66} modes considered in Fig. 3, where **H** is along (1,1,0), the equilibrium-strain contributions to $\Delta v/v$ are again rather small, but the correction given by (35) is of decisive importance for the difference between the field dependence of the two velocities. The sign of the splitting is reversed in comparison with Fig. 2. This change of sign only applies to the rotational contributions, which are here about twice as large as the dipolar contributions.

V. CONCLUSION

TmSb is a simple magnetic system. The crystal-field symmetry is cubic, and the ground state is a singlet with no signs of any significant modifications due to two-ion interactions. This makes the system suitable for a sensitive test of the magnetoelastic effects predicted from the requirement of rotational invariance. We have reconsidered the theory for these effects. The rotational interactions only influence the sound velocities in the presence of an external magnetic field, independently of the crystal symmetry. This is shown by performing a unitary transformation of the Hamiltonian, which also leads to simplifications in the account of the low-frequency rotational effects. Contrary to the previous analyses we find that the magnetoelastic second-order dynamic effects on the acoustic sound waves cancel out, within the harmonic

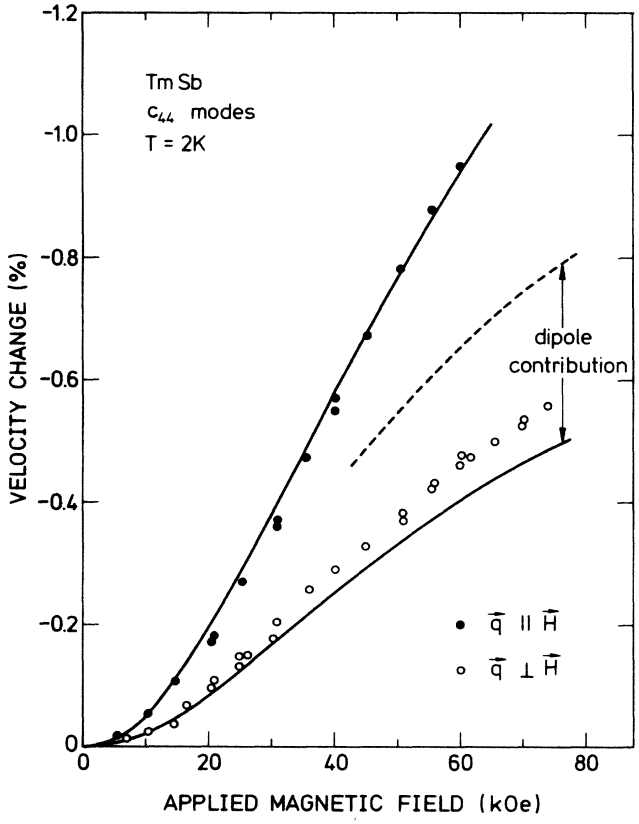

FIG. 2. The relative (nominal) velocity changes of the c_{44} modes in TmSb at $T=2$ K, as functions of an applied magnetic field along (1,0,0). The experimental results are taken from Wang and Lüthi (Ref. 9). The solid circles denote the case where the propagation vector **q** of the transverse sound waves is parallel to **H**, and the open circles show the changes when **q** is perpendicular to **H** and the polarization vector is parallel to **H**. The solid lines show the theoretical results obtained from Eq. (30) using $G_3=1.85$ meV together with the other parameters given in the text. The contribution due to the dipole interaction is indicated on the figure.

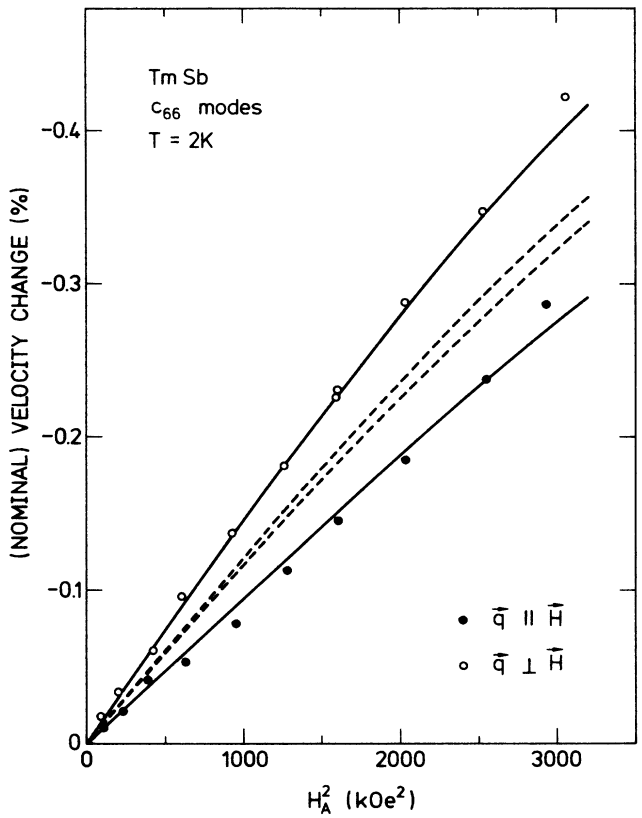

FIG. 3. The nominal relative velocity changes of the c_{66} modes in TmSb at 2 K as functions of the square of the external magnetic field H_A applied along (1,1,0). The experimental results are determined by Wang and Lüthi (Ref. 9). The polarization vector of the transverse sound waves is along $(1,-1,0)$ when $\mathbf{q}\|\mathbf{H}$ (solid circles), and the other mode is the one obtained when interchanging the directions of the propagation and polarization vectors. The theoretical predictions shown by the solid lines are obtained from Eq. (31) with $G_2=-9.60$ meV, and they include the corrections (35) to the actual velocity changes, calculated using $G_3=1.85$ meV. The dashed lines show the calculated results for the actual velocity changes. The upper and lower dashed lines are associated, respectively, with the upper and lower solid lines.

approximation, due to the equilibrium conditions. Instead we account for the effects of the nonzero equilibrium strains, assuming the elastic Hamiltonian to be quadratic and the magnetoelastic Hamiltonian to be linear in the deformation matrix $\overleftrightarrow{E}$. These effects are of importance in principle, even though the modifications in TmSb are only of the order of 1–4%. If the strain-dependent corrections were significant the higher-order strain terms in the Hamiltonian should also be considered, especially the third-order elastic constants which contribute to $\Delta c/c$ with terms of the order of the equilibrium strains. In the case of TmSb, Wang and Lüthi[9] did estimate the importance of the magnetoelastic couplings quadratic in the strains, as well as the linear ones involving octupole operators, by utilizing the point-charge model, and they concluded that these terms should have no serious consequences.

The agreement between the calculations and the experimental field dependences of the transverse sound-wave velocities in TmSb is very close. This leaves very little room for possible effects of the third-order elastic constants, in accordance with the fact that the third-order modification of the velocity of a transverse sound wave is normally small when volume changes can be neglected. The only uncertainty is in the temperature dependences of the nonmagnetic elastic constants which could lead to a 5–10% change of G_2 or G_3. These uncertainties have nearly no influence on the calculated differences in the velocities found on interchanging the directions of propagation and the polarization vectors for the transverse sound waves. These differences observed in TmSb by Wang and Lüthi[9] are here reproduced quite accurately with no adjustable parameters. The explanation involves both the interaction terms introduced when the total system is required to be rotationally invariant, and the rapid variation of the magnetic dipole-dipole coupling occurring at long wavelengths. The two mechanisms are of roughly equal importance.

The present theory can be generalized straightforwardly to cover other cases. We have applied the theory to SmSb, which closely resembles TmSb, and using the parameters deduced by Mullen *et al.*,[17] we find the field dependences of the c_{66} modes to be vanishingly small in accordance with the observation of Mullen *et al.* (at 6.7 K, where the presence of the magnetic phase transition at $T_N=2.1$ K can be neglected). In the case of $CeAl_2$ the rotational interactions and, in addition, the dipolar interactions only lead to small effects, in agreement with the experiments and the numerical analysis of Lüthi and Lingner.[19] We have also considered the basal-plane ferromagnet Tb metal (hexagonal closed packed), analyzed[14] previously estimating the rotational effects to be unimportant. This approximation is justified; the corrections are proportional to the internal magnetic field and they modify the field dependence of c_{66} only by a few percent. These corrections are nearly of the same magnitude as those produced by the third-order elastic constants, using the values calculated by Rao and Ramanand.[20] One interesting phenomenon is worth mentioning, namely that the rotational interactions produce a slight shift in the critical field H_c, which is required to rotate the moments to be along a hard a axis. A shift of H_c may be introduced by the external mechanical forces, which are needed in order to prevent the crystal from rotating. With the appropriate boundary conditions, the change in H_c is found to compensate the effects of the rotational interactions, so that c_{66} (with $\mathbf{q}\|\mathbf{H}$) still vanishes at the critical field.

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