

## Field dependence of the elastic constant $c_{66}$ in the basal-plane ferromagnet terbium

J Jensen† and S B Palmer‡

† Physics Laboratory I, HC Ørsted Institute, University of Copenhagen, Denmark

‡ Department of Applied Physics, University of Hull, Hull HU6 7RX, UK

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**Abstract.** The dependence of  $c_{66}$  in ferromagnetic Tb on a magnetic field applied along a hard axis in the basal plane has been studied experimentally. It has been predicted that  $c_{66}$  should vanish at the second-order transition from a phase where the magnetisation is parallel to the field to a phase which has a finite moment perpendicular to the field. Although it has not been possible to detect the ultrasonic waves in the close neighbourhood of the transition,  $c_{66}$  is found to be substantially reduced at fields slightly higher than the critical value. The observed behaviour of  $c_{66}$  can be described quite accurately, at all temperatures below the Curie point, by a random-phase theory without the use of any free parameters.

### 1. Introduction

The strong coupling between the lattice and the spin system which has been observed in many of the rare-earth metals, particularly Tb (Rhyne and Legvold 1965), leads to a number of phenomena. For an easy planar magnet like Tb those magnetoelastic couplings which might distort the hexagonal symmetry of the basal plane are of fundamental importance. As discussed by Cooper (1968), this applies to the transition at  $T_c$  from the helically ordered to the ferromagnetic phase ( $T_c$  is about 220 K in Tb). These  $\gamma$  strain couplings also provide the dominating contribution to that part of the free energy which determines the easy axis within the basal plane (Houmann *et al* 1975). In zero field and below  $T_c$ , Tb is ferromagnetic with the moments lying in the basal plane along a  $b$  axis. By the application of a magnetic field of an accessible magnitude (32 kOe) at low temperatures along an  $a$  axis, the moments can be aligned in this direction. Below the critical field the magnetisation makes a finite angle  $\phi$  with the  $a$  axis, and this angle vanishes for an applied field larger than the critical value. Unless very strong renormalisation effects are present, the transition between the two phases is of second order (see Jensen 1975). The second derivative of the free energy with respect to  $\phi$ ,  $F_{\phi\phi}$ , vanishes at the transition, which implies that the transverse susceptibility at zero frequency and wavevector diverges at the critical field. If the ferromagnetic spin system were isolated, this divergence would correspond to a zero-frequency spin-wave excitation at zero wavevector, analogous to an easy planar magnet without any anisotropy in the hexagonal plane.

However, as first discussed by Turov and Shavrov (1965), the coupling of the spin system to the  $\gamma$  strains modifies the dynamic behaviour in such a way that the energy

gap in the spin-wave spectrum stays finite at the transition. In their model, Turov and Shavrov neglected the dynamics of the lattice, which led them to name it the 'frozen lattice' model. Later, the dynamics of the combined spin-lattice system were analysed in more detail by several authors (Jensen 1971a, b, Liu 1972, Chow and Keffer 1973). They found that the frozen lattice model gave a correct description of the energy gap, because essentially the frequencies of the long-wavelength phonons are zero compared with the spin-wave energies. The divergence of the transverse susceptibility is connected instead with a soft mode in the phonon spectrum, in the sense that the velocity of the transverse sound waves, propagating and polarised in the basal plane, goes to zero at the transition. In fact the normal modes of the coupled system are no longer pure magnons or phonons since the wavefunctions are mixed. However, in the ultrasonic regime considered here, this mixing is extremely small. This is in many respects similar to what occurs at the structural phase transition in a cooperative Jahn-Teller system like  $\text{TbVO}_4$  or  $\text{DyVO}_4$  (Sandercock *et al* 1972).

In the following section we outline the theoretical considerations and in §3 we present the experimental results for the transverse sound velocities, corresponding to the elastic constant  $c_{66}$ , for the waves propagating either parallel or perpendicular to the applied field. The degeneracy of the two kinds of modes is lifted because of the long-range magnetic dipole forces. A comparison between the random-phase theory and experiments is given and we finally summarise and discuss the data in the last section.

## 2. Theory

The random-phase theory (RPA) for the coupled spin-lattice system in the ferromagnetic phase of Tb has been worked out by a number of authors (Jensen 1971a, b, Liu 1972, Chow and Keffer 1973), each using slightly different techniques. We will not give the details here, but will quote the results that are of interest in the present context.

The equilibrium values of the homogeneous  $\gamma$  strains are given in terms of the two magnetostriction coefficients  $C$  and  $A$  (see Rhyne and Legvold 1965):

$$\frac{1}{2}(\bar{\epsilon}_{11} - \bar{\epsilon}_{22}) = C \cos 2\phi - \frac{1}{2}A \cos 4\phi \quad (1a)$$

$$\bar{\epsilon}_{12} = C \sin 2\phi + \frac{1}{2}A \sin 4\phi. \quad (1b)$$

The magnetisation is assumed to be perpendicular to the  $c$  axis and  $\phi$  is the angle between the direction of magnetisation and the  $a$  axis, parallel to the 1 axis.  $C$  and  $A$  generally depend on both temperature and field. According to the renormalisation theory described by Callen and Callen (1965), it is possible to express the dependence of  $C$  and  $A$  on the state variables through their dependence on the relative magnetisation,  $\sigma = \sigma(T, H)$ . For instance  $C = C(T, H) = C(\sigma)$  and if  $C$  is assumed to be of lowest-order single-ion origin, then  $C(\sigma) \propto \hat{I}_{5/2}(\sigma)$ . We will not make use of the last relation; instead we use directly the experimental results described by Rhyne and Legvold (1965), which, however, show good agreement with the theory of Callen and Callen.

The temperature dependence of  $\sigma$  in zero field is taken from the magnetisation curves measured by Hegland *et al* (1963) and at finite field we use

$$\sigma(T, H) = \sigma(T, 0) + (d\sigma/dH) H_i \quad (2a)$$

where the molecular-field value used for  $d\sigma/dH$  is

$$\begin{aligned} \frac{d\sigma}{dH} &= g\mu_B J [1 - \sigma(T, O)] / k_B T_c \\ &= 0.0027 [1 - \sigma(T, O)] \text{ kOe}^{-1} \end{aligned} \quad (2b)$$

where the various symbols have their usual meaning.  $H'_i$  is the internal field  $H_i$  if applied along an easy axis. If the field is applied along an  $a$  axis, we further subtract the critical field  $H_c$ , i.e. we neglect possible variation of  $\sigma$  with the angle  $\phi$ . This procedure gives a fair account of the magnetisation curves (Hegland *et al* 1963, P Touborg private communication) and also of the forced (field-dependent) magnetostriction as shown by Houmann *et al* (1975), at least for  $\sigma$  larger than 0.7.

If a field is applied along the hard planar axis, then the magnetisation is pulled towards this direction, as described by the equation

$$H_i = \frac{1}{6} H_c \sin 6\phi / \sin \phi \quad (3a)$$

where in terms of the applied field  $H$

$$H_i = H - H_a \cos \phi \quad H_a = N_{\perp} M. \quad (3b)$$

$M$  is the magnetisation and  $N_{\perp}$  is the demagnetisation factor perpendicular to the  $c$  axis (the crystal is assumed to be a disc with its axis along the  $c$  axis). Equation (3) determines  $\phi$  as long as  $H_i$  is positive, that is  $H > H_a \sqrt{3}/2$ , but smaller than  $H_c$ . When  $H_i$  is equal to or larger than  $H_c$ ,  $\phi$  is zero. The critical field  $H_c$  is determined by two contributions:

$$H_c = (36K_6^6 + 72c_{66}^0 AC) / M. \quad (4)$$

The first is the pure magnetic anisotropy and the second arises from the magnetoelastic deformation of the basal plane.  $H_c$  has been measured by several methods and is known quite precisely to be equal to  $32.3 \sigma^{1.5}$  kOe (Houmann *et al* 1975).  $c_{66}^0$  is the unperturbed value of the elastic constant.

If we define

$$\Gamma = 16c_{66}^0 (C^2 + A^2 + 2AC \cos 6\phi) \quad (5a)$$

and

$$G_q^{-1} = M(H_i \cos \phi - H_c \cos 6\phi + H_M) + D\sigma^2 q^2 \quad (5b)$$

then the elastic constant  $c_{66}$ , relative to the unperturbed value, is given by (Jensen 1971a, b, Liu 1972, Chow and Keffer 1973):

$$c_{66}/c_{66}^0 = (\omega(\mathbf{q})/\omega_0(\mathbf{q}))^2 = 1 - [\Gamma/(G_q^{-1} + \Gamma)] = 1/(1 + \Gamma G_q). \quad (5c)$$

$c_{66}$  can be obtained from the velocity  $\omega(\mathbf{q})/q$  of the transverse phonons, polarised and propagating in the basal plane and  $H_M$  is the contribution from the magnetic dipole coupling.

Because of the long-range nature of this coupling,  $H_M$  changes rapidly with the wave-vector so that

$$H_M = \begin{cases} 4\pi M \sin^2(\phi - \psi) & q \gg 2\pi/L \\ N_{\perp} M \cos^2 \phi & q \approx 2\pi/L. \end{cases} \quad (5d)$$

$\psi$  is the angle between the wavevector and the 1 axis and  $L$  is the diameter of the disc (see the discussion by Keffer 1966).  $D$  in equation (5b) is the stiffness constant of the magnetic two-ion coupling. In the ultrasonic experiment reported in the next section, the wavelength of the ultrasonic waves considered is of the order of 0.1 mm. This means that the last term in equation (5b) is extremely small and it is included here only to indicate that the dispersion relation of the transverse phonons  $\omega(\mathbf{q})$  becomes quadratic in  $q$  if the first bracket in equation (5b) vanishes. The crystal diameter is of the order of a few mm, which implies that  $q \gg 2\pi/L$ .  $4\pi M$  is equal to  $34\sigma$  kOe (Hegland *et al* 1963), which compares with  $H_c$  at low temperatures and we notice that this term removes the degeneracy of the transverse modes which propagate either parallel or perpendicular to the magnetisation.

In equation (5) we have neglected a number of effects which are all very small. Firstly, the longitudinal and transverse phonons become indirectly coupled via the spin waves if the magnetisation is not along a symmetry direction,  $\phi \neq p(\pi/6)$ , or if the wavevector is not parallel or perpendicular to the magnetisation,  $\phi - \psi \neq p'\pi/2$  ( $p$  and  $p'$  are integers). Secondly, we have not included the term arising from the requirement of rotational invariance of the spin-lattice interaction (see for instance Dohm and Fulde 1975), as it is entirely dominated by the corresponding effect of  $H_M$ . Finally, we neglect higher-order changes in the elastic constant  $c_{66}^0$ , caused by changes in the lattice parameters, which have been reported to be present in the paramagnetic phase of Tb (Torikachvili *et al* 1979).

As mentioned above, the result given by equation (5) has been deduced by several authors, all using essentially an RPA decoupling of the coupled equations of motion. By this procedure it is in fact the spin-wave energy parameter  $A(\mathbf{q}) - B(\mathbf{q})$  defined by Houmann *et al* (1975), which appears in the denominator of equation (5c) rather than the corresponding factor  $g\mu_B(G_q^{-1} + \Gamma)/M$ . Houmann *et al* determined a difference between the two quantities by comparing their energy gap parameters with the static values that appear in equations (5a) and (5b). As discussed by Jensen (1975), these differences may be produced by renormalisation effects of higher order than RPA or by coupling to the polarised conduction electrons. In both cases the contribution will scale with the frequency, which implies that in the ultrasonic regime the static parameters are more appropriate than those deduced at the much higher frequencies of the spin waves.

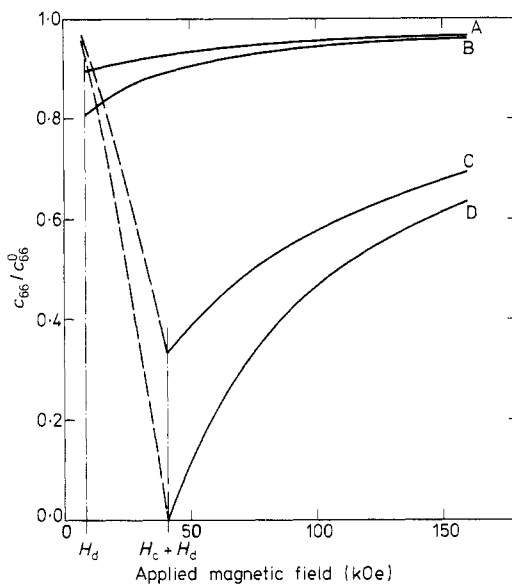
The finite-temperature result given by equation (5) is the same as that obtained by a simple generalisation of the zero-temperature result. The generalisation is justified to first order in temperature by performing the calculation to next order beyond the harmonic boson approximation in the expansion produced by the generalised Holstein-Primakoff transformation (see for instance Jensen 1975, also Brooks and Egami 1974). The same result may also be obtained by considering the second derivative of the free energy with respect to the strain  $\epsilon_{12}$ . This method gives essentially the same answer, independent of the specific form of the Hamiltonian. The result (5) does not depend on whether the anisotropy  $K_6^0$  or the magnetostriction  $A$  and  $C$  originate from single-ion crystalline-field effects, as is normally assumed in the calculations, or whether anisotropic two-ion couplings are contributing (see also the discussion by Houmann *et al* 1975).

When  $\phi = 0$ ,  $H_i \geq H_c$ , the transverse susceptibility at zero frequency and wavevector is proportional to the inverse of the second derivative of the free energy with respect to  $\phi$ ,  $F_{\phi\phi}^{-1} = G_{q=0}$ , which is valid at least as long as the spin waves may be considered to be non-interacting bosons (see the discussion by Jensen 1975).  $G_{q \approx 0}$  given by equations (5b) and (5d) diverges at  $H_i = H_c$  except for the dipole contribution

$H_M$ . In the extreme long-wavelength limit,  $H_M$  is finite. However, this finite-size effect is not present when  $q \gg 2\pi/L$  and the energy of the transverse phonon mode that propagates parallel to the magnetisation, see equation (5c), should vanish. If the temperature is not too close to the Néel temperature,  $T_N \approx 230$  K, the critical fluctuations should be unimportant. This may be seen by applying the real-space criterion given by Als-Nielsen and Birgenau (1977). If  $H_M$  is neglected then  $G_q$  is proportional to a mean-field susceptibility for a system of marginal dimensionality  $d^* = 3$ , which corresponds to  $c_{44}$  being the soft elastic constant in a crystal of cubic symmetry at a structural phase transition (Cowley 1976). The presence of  $H_M$ , neglecting the finite-size effect, further reduces the importance of the critical fluctuations as  $d^*$  now becomes equal to 2, which implies that the RPA expression (5) should also be applicable close to the critical field.

Above the critical field only inelastic magnetic processes contribute to the transverse phonon energies and to a first approximation, the contribution of the spin waves to the damping coefficient of the ultrasonic waves is extremely small. At all temperatures where Tb is ferromagnetic, the spin waves at long wavelengths are well defined; the intrinsic linewidth is of the order of one tenth of the frequency (Nielsen *et al* 1970). This number is divided by a factor of about  $10^6$ , namely the ratio between the frequencies of the spin waves and the ultrasonic waves in the expression for the damping coefficient. Instead, other contributions to the damping, related to intrinsic properties of the phonon system of the metal, might be of importance, since they increase as  $1/c_{66}$  (at constant frequency) when  $c_{66}$  softens.

Figure 1 shows the behaviour of  $c_{66}/c_{66}^0$  predicted by equation (5) for Tb at zero temperature, when a field is applied along an easy or a hard planar axis. For the parameters that appear in the equations above, we use the experimental values given in table 1. As illustrated by the figure, we expect a strong dependence of  $c_{66}$  on field when the



**Figure 1.** The field dependence of  $c_{66}/c_{66}^0$  at zero temperature for Tb as predicted by the parameters used in table 1, using  $c_{66}^0 = 2.88 \times 10^{10}$  N m<sup>-2</sup>. The four curves A–D correspond to determination of  $c_{66}$  from ultrasonic waves propagating parallel, curves B and D, or perpendicular, curves A and C, to the field. The field is applied along the easy axis for curves A and B and along the hard  $a$  axis for curves C and D.

field is applied along the hard axis and a more moderate dependence when application is along the easy axis. The difference between the two cases in which the wavevector of the ultrasonic waves is parallel or perpendicular to the magnetisation should also be quite substantial. The broken curves show the behaviour expected for  $c_{66}$  in magnetic fields smaller than  $H_c + H_d$ , applied parallel to the  $a$  direction. It is evident that walls

**Table 1.** Values and magnetisation exponents of the different parameters which enter the expression for the relative elastic constant,  $c_{66}/c_{66}^0$ , in Tb. The magnetisation exponents of  $C$  and  $A$  (and presumably also  $H_c$ ) only apply for  $\sigma$  larger than 0.75.  $C$  is the mean value of the two experimental results of Rhyne and Legvold (1965).

Parameter	Value	Exponent
$C(\times 10^3)^a$	4.20	2.7
$A(\times 10^3)^a$	2.10	8.0
$H_c(\text{kOe})^b$	32.3	15.0
$4\pi M(\text{kOe})^c$	34.0	1
$H_d(\text{kOe})$	8.5	1

<sup>a</sup> Rhyne and Legvold (1965).

<sup>b</sup> Houmann *et al* (1975).

<sup>c</sup> Hegland *et al* (1963).

between the two kinds of domains, in which the magnetisation makes the angle  $\pm\phi$  with the  $a$  axis, may well have an appreciable effect on the ultrasonic waves. When the applied field is smaller than the demagnetising field, the direction of magnetisation in the domains is distributed among all the different  $b$  axes. This will give rise to a similar distribution of magnetic contributions to  $c_{66}$ , which would show up as a strong damping of the ultrasonic waves, in accordance with the observations of Palmer *et al* (1974).

### 3. Experiments

The elastic constants of Tb have been reported by Palmer *et al* (1974) for the temperature range 0–300 K. The experiments were performed both at zero field and with a field of 25 kOe applied along an easy axis. Here we supplement these measurements with a detailed study of the behaviour of  $c_{66}$  when the field is applied along a hard  $a$  axis. The results presented here and previously were obtained with a single crystal grown by Metals Research Ltd using a zone flotation technique. The sample was initially in the form of a disc with its axis along the  $c$  direction, but to allow measurements of  $c_{66}$ , two plane-parallel faces were cut normal to a basal plane axis. The original disc was about 6 mm diameter by 3 mm thick and the separation of the faces parallel to the  $c$  axis was approximately 3.5 mm. Allowing for the plane-parallel faces, which are needed for the ultrasonic measurements, the demagnetisation field in the basal plane  $H_d$  should be reasonably small and homogeneous. In the analysis, the precise value of  $H_d$  is not of great importance and in all cases we use  $H_d \simeq 4\pi M/4 = 8.5\sigma\text{kOe}$ , as estimated from the geometry of the disc.

The ultrasonic waves were generated and detected by 15 MHz quartz transducers that were bonded to the two plane-parallel faces with Araldite epoxy resin. The pulse-echo overlap technique was used to measure the acoustic velocity (Papadakis 1964).

Sample temperatures were kept stable to better than 0.1 K using a commercial temperature controller; temperature measurement with a 0.3% Fe–Au/v Chromel thermocouple gave an absolute accuracy of  $\pm 0.5$  K. Magnetic fields were generated with both an 11 inch electromagnet and a superconducting magnet.

Corrections were not made for changes in path length or density caused by thermal expansion and magnetostriction. These are orders of magnitude smaller than the effects reported here. The density used to calculate the elastic constant from the measured acoustic velocity was  $7895.4 \text{ kg m}^{-3}$  (Spedding 1971). It is likely that the overall purity of the sample is not better than 99% but measurements on samples of higher purity from the Centre for Materials Science, Birmingham University, are planned for the near future.

According to the theory discussed in the previous section, we should expect to observe reasonably welldefined ultrasonic waves even when their velocity approaches zero, as has been found for the analogous systems TbVO<sub>4</sub> and DyVO<sub>4</sub> (Sandercock *et al* 1972). However, it was only possible to detect the transmitted ultrasonic signals if the corresponding elastic constant was larger than approximately half the unperturbed value. Contrary to the Jahn–Teller systems, the dimension of the Tb crystal changes rapidly when cooled, because of the large magnetostriction present below the magnetic ordering temperature  $T_N$ . These deformations make it very hard to maintain acoustic contact with the sample (see the discussion by Palmer 1976). It therefore proves very difficult to separate the inherent ultrasonic attenuation associated with the softening of the  $c_{66}$  mode from the attenuation produced by strains in the transducer–bond–sample region. As the velocity falls, the corresponding decrease in the acoustic signal makes the effects of the bond more and more important. In an attempt to overcome the bond problem, we tried to replace the quartz transducer by thin-film CdS transducers. However, we here encountered a number of other problems which have not yet been solved. In particular, the definition of the shear vibration direction for CdS transducers is  $\pm 5^\circ$  and this varies across a particular transducer, which to some extent averages out the softening of  $c_{66}$ . All the experimental results to be presented here were obtained with the quartz transducers. Because of the bond problem, these measurements have required a great number of experimental runs (and a number of results which were not reproducible has been discarded).

The only one of the parameters that appears in equation (5) and which is not determined by other experiments is the unperturbed elastic constant  $c_{66}^0$ . In order to obtain a precise determination of  $c_{66}^0$ , we include the results of Palmer *et al* (1974) for  $c_{66}$  as a function of  $T$  in an applied field of 25 kOe along an easy axis. These results are shown in figure 2, together with our results for  $c_{66}$  when a field of 20 kOe is applied along the  $a$  axis. These latter measurements are for ultrasonic waves propagating both parallel and perpendicular to the magnetisation. With the help of equation (5), the three different cases lead to a fairly accurate and reasonable result for  $c_{66}^0$  as a function of  $T$ , which is shown by the broken line in figure 2. The introduction of this result for  $c_{66}^0$  into equation (5) produces the results shown by the full curves in figure 2, which account for all the experimental results within a few per cent.

Figure 3 shows  $c_{66}$  at various temperatures as a function of field applied along the  $a$  axis, determined by the velocity of the ultrasonic waves propagating perpendicular to the field. In this configuration  $c_{66}$  should exhibit a minimum at the transition when  $H$  is equal to  $H_c + H_d$ ; the broken curve in figure 3 gives the calculated value of  $c_{66}$  at this field. Except at the highest temperatures, there are practically no experimental results on the low-field side of the broken curve, because of the very high ultrasonic

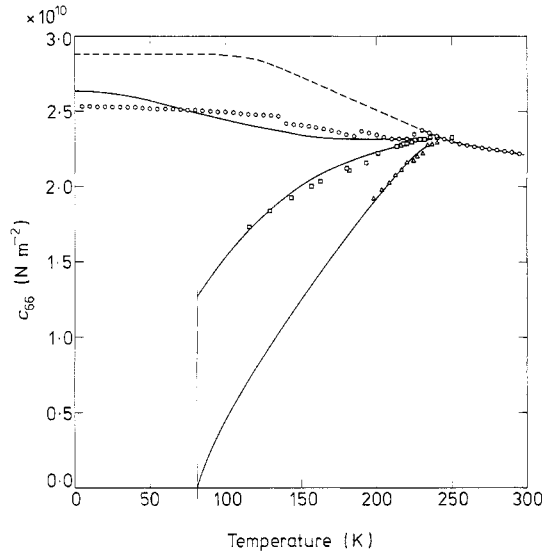


Figure 2. The temperature dependence of  $c_{66}$  in Tb with  $\circ$ ,  $H = 25 \text{ kOe} \parallel b \text{ axis}$ ,  $q \perp H$  (after Palmer *et al* 1974);  $\square$ ,  $H = 20 \text{ kOe} \parallel a \text{ axis}$ ,  $q \perp H$ ;  $\triangle$ ,  $H = 20 \text{ kOe} \parallel a \text{ axis}$ ,  $q \parallel H$ .  $H$  is the applied field and  $q$  is the wavevector of the ultrasonic waves. The broken curve is the predicted value of  $c_{66}^0$  and the full curves (below 230 K) are the theoretical results for the three sets of conditions.

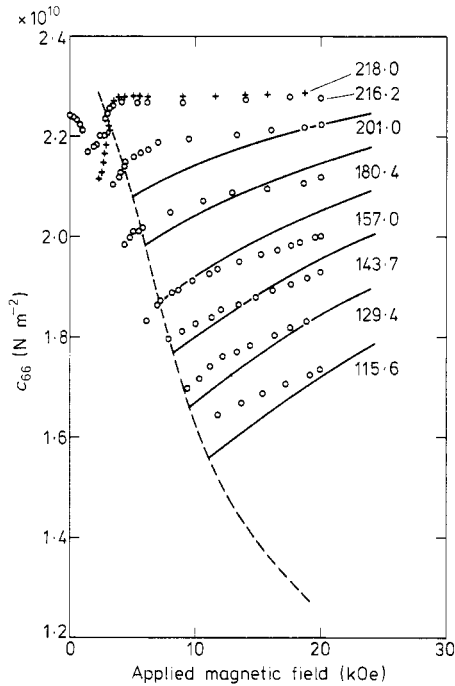
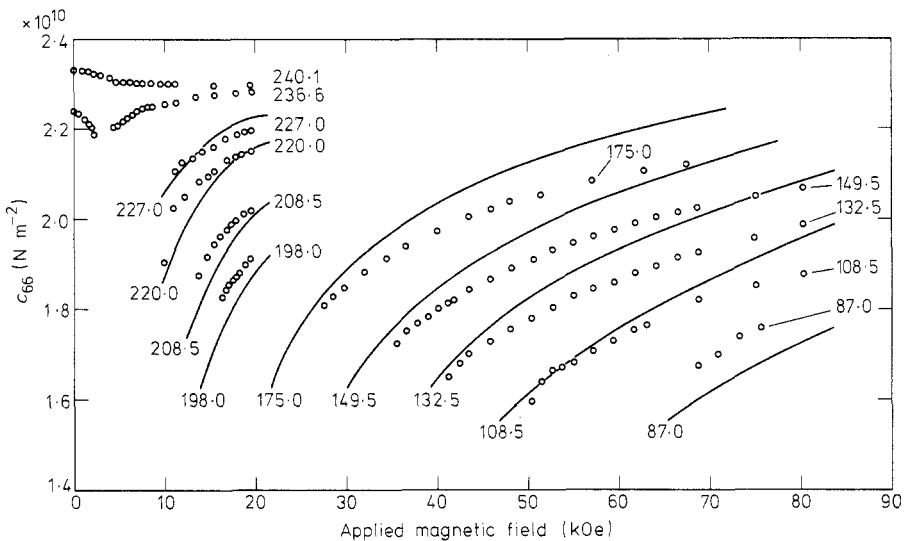


Figure 3. The results for  $c_{66}$  at various temperatures (K) in Tb as function of the field applied along the  $z$  axis with  $q$  perpendicular to the field. The broken curve shows the calculated value of  $c_{66}$  when the applied field  $H$  is equal to  $H_c + H_d$ . The full curves display the field dependences of  $c_{66}$  predicted for the various temperatures.



attenuation in this region. We should add that the acoustic bondings may be strongly affected by the great variation of the lattice parameters that occurs just below the critical field where  $\phi$  becomes non-zero (see equation 1). At the highest temperatures ( $\sigma$  less than about 0.6)  $H_M$  is the dominant quantity in the expression for  $G_q$  (equation 5b) and because  $H_M$  decreases when  $H$  becomes smaller than  $H_d (\gg H_c)$ , but larger than  $H_d\sqrt{3}/2$ , the minimum at the transition is eliminated in accordance with the observations above 200 K. The minimum at 216.2 K occurs at a field  $H \approx H_d/2$ , where the crystal is divided into several different domains leading to a somewhat fortuitous value of  $c_{66}$ . The full curves in figure 3 display the results predicted by equation (5).  $c_{66}$  has not been calculated at the highest temperatures, because here the low-field values of  $\sigma$  are too uncertain. The zero-field value of  $\sigma$  in equation (2a) is determined by an extrapolation of the linear part of the magnetisation curves, which corresponds to the use of a linear dependence of  $\sigma$  with field in this equation. However, when  $\sigma$  is small (less than about 0.6), this procedure overestimates the value of  $\sigma$  at low fields. This uncertainty in  $\sigma$  is presumably the most important reason for the discrepancy that appears between the calculated and experimental results at 201 K.



**Figure 4.**  $c_{66}$  in Tb at various temperatures (K) determined from the ultrasonic waves propagating parallel to the field applied along the hard  $a$  axis. The full curves show the calculated soft-mode behaviour of  $c_{66}$  at the various temperatures. A few examples of the field dependence of  $c_{66}$  observed above  $T_N$  are also given.

Finally, figure 4 gives the results obtained for  $c_{66}$  when the ultrasonic waves propagate parallel to a field applied in the  $a$  direction. The experimental results show a clear tendency towards the soft-mode behaviour predicted by the theory and shown by the full curves. Unfortunately, it was not possible to measure  $c_{66}$  down to its minimum value because of very high attenuation, which might be inherent or may be because of the effect of bond strain as discussed above. The high-field measurements reveal a systematic deviation between theory and experiment which seemingly increases as the square of the field. This discrepancy is surprising, when related to the good representation of the absolute changes predicted by the theory. For instance, the small contributions which have been neglected as discussed in the preceding section might give rise to corrections of the

order of a few per cent, but these should be fairly independent of field. As discussed in §2 the calculations include the effect of the field dependence of  $\sigma$  and thereby of all the other quantities that appear in equation (5). These changes in the parameters are moderate and should be well represented by the procedure used, at least for  $\sigma$  larger than 0.7 ( $\sim 175$  K). The distortion of the ultrasonic signals produced by incomplete acoustic contact with the sample might give rise to the systematic deviations observed, which demands some caution in the interpretation of the discrepancy.

#### 4. Discussion

The dependence of the elastic constant  $c_{66}$  in ferromagnetic Tb on a magnetic field applied in the basal plane has been studied in detail. At the wavelength considered, the degeneracy of the transverse ultrasonic waves, propagating and polarised in the basal plane, is observed to be lifted. We have explained the lifting by the presence of long-range magnetic dipole coupling. Clear evidence is presented for a softening of the long-wavelength phonons propagating parallel to the magnetisation. The difficulties in maintaining acoustic contact with the sample prevented us from measuring  $c_{66}$  close to the critical field  $H_c$  of the second-order transition. However, reductions of the order of 50% were observed.

The random-phase theory describing the field dependence of  $c_{66}$  was carefully examined. The critical fluctuations close to the phase transition at  $H_c$  were argued to be unimportant, as the marginal dimensionality was found to be two. Damping effects caused by the coupling to the spin waves should be negligible and other contributions arising from phonon-phonon and phonon-electron interactions should only be considerable when  $c_{66}$  is close to zero. The result given by equation (5) should be applicable within a wide range of values of the magnetisation and should be fairly independent of the model considered. We might add that the renormalisation effects produced by the strong axial anisotropy of Tb (see for instance Jensen 1975) does not affect the result to leading order in the anisotropy.

The comparison between theory and experiment, in which  $\sigma$  ranges from 0.9 to 0.5, is very satisfactory, especially when considering that we use essentially no free parameters. The only parameter not given beforehand was the 'non-magnetic' value of  $c_{66}$ , i.e.  $c_{66}^0$ , but this could only be varied within a very narrow range and could be estimated from the easy-axis results of Palmer *et al* (1974). The only discrepancies detected are the deviation between the theoretical and experimental high-field slopes in figure 4 and the occurrence of strong damping effects close to the critical field. Both discrepancies might be produced by damage in the acoustic contact to the sample and certainly this possibility cannot be excluded without further experimental evidence.

The overall good agreement between calculation and the experiment yields an independent check (with an accuracy of about 5% for  $C + A$ ) of the magnetostriction coefficients  $C$  and  $A$  measured by Rhyne and Legvold (1965), and can be compared with the discussion given by Martin and Rhyne (1977). Also, it is clearly not the corresponding spin-wave energy parameter that appears in equation (5), in accordance with the discussion in §2. The value deduced for the unperturbed elastic constant at zero temperature,  $c_{66}^0 = 2.88 \times 10^{10} \text{ N m}^{-2}$ , is only slightly larger than that used by Houmann *et al* (1975) and the magnetoelastic contribution to  $H_c$  in equation (4) is determined to be 68 kOe at zero temperature, which is approximately twice the final value of  $H_c$ .

The experimental difficulties in measuring the velocity of ultrasonic waves in crystals with large magnetostriction are severe. We hope to be able to improve the technique described here in order to get more definitive results; the most promising possibility is the use of thin-film CdS transducers (Palmer 1976). It should be of interest to study the soft-mode behaviour closer to the transition and to determine the importance of the intrinsic damping of the ultrasonic waves. It would be of interest to perform a similar study on Dy. Although the behaviour of  $c_{66}$  in Dy should be very similar to that in Tb, there are two differences which might be of importance. Firstly  $A$  is close to zero in Dy (Martin and Rhyne 1977) and hence  $H_c$  is determined entirely by the magnetic anisotropy  $K_6^6$ ; secondly the renormalisation effects attributable to the magnetic anisotropy are more important in Dy than in Tb, mainly because  $H_c$  is a factor of four larger than in Tb (Jensen 1975).

The random-phase theory in §2 gives an adequate description of the behaviour of  $c_{66}$  in ferromagnetic Tb. The corresponding result for the energy of the long-wavelength spin waves is also in accordance with experiment (Houmann *et al* 1975). We may therefore conclude that the dynamics of the spin-lattice system are well understood, although which mechanism is responsible for the magnetic absorption peak close to the transition at  $H_c$ , observed by for instance Hart and Stanford (1971), is still not clear. The measuring frequency used by these authors, 24 GHz, is small compared with the spin-wave frequencies. The effect attributable to the magnetic part of the low-energy modes, which are for the main part phonon-like, seems to be very small as discussed by Liu (1972). As suggested previously (Jensen 1971a, b) it is very likely that the dynamic effects of the domain walls, which are present when the field is just below the critical value, must be included to obtain a complete description of the phase transition.

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