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Magnetoelastic effects in single-crystal erbium

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Abstract. We have measured the ultrasonic elastic constants C_{33}, C_{44}, C_{11} and C_{66} and their associated attenuation coefficients (α_{ij}) for a single crystal of erbium as a function of temperature. We have observed anomalous behaviour in both C_{ij} and α_{ij} which we attribute to commensurate spin-slip structures as observed by Gibbs *et al* and Lin *et al* using synchrotron x-ray scattering and neutron diffraction techniques.

1. Introduction

The magnetic structure of erbium has been the subject of extensive neutron scattering studies [1–3], which have identified three principal phases of magnetic order.

Below a Néel temperature (T_N) of approximately 84 K the spins adopt a sinusoidally modulated structure with the magnetization confined parallel to the c-axis (the hexagonal axis). The period of the modulation decreases from just under 8 atomic layers at 83 K down to less than 7 atomic layers at approximately 53 K. At 53 K (T_m), a basal plane component of the magnetization develops and is modulated with the same period as the c-axis component. The period of the modulation increases until it appears to lock in to an eight-layer structure at 21 K. At 18 K (T_c) the c-axis component becomes ferromagnetic producing a low temperature ferro cone structure via a first order transition. Below 18 K the modulation of the basal plane component locks in to an incommensurate value of 8.4 atomic layers.

It is the intermediate phase, between 18 K and 53 K, which has proved to be the most difficult to identify definitively. The most widely accepted model is that of Jensen [4] in which the spins trace out an ellipse in the a–c plane when consecutive spins are drawn from a common origin. The c-axis component of the magnetization is clamped to the basal plane component in such a way that when one is at a maximum the other is at a minimum. It is however, the x-ray scattering work of Gibbs *et al* [5] which has provided most of the detailed structure of the intermediate magnetic phase of erbium.

Higher-order scattering peaks on the neutron data of Habenschuss *et al* [3] have shown the gradual squaring up of the c-axis modulation with decreasing temperature; however, Gibbs *et al* showed that this squaring up of the modulation is accompanied by the formation of several commensurable structures, some of which exhibit net ferromagnetic (strictly ferrimagnetic) components and are thus stabilized by the magnetoelastic energy and exist over temperature ranges of two or three Kelvin.

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Each of these commensurate structures are made up of blocks of four atomic layers (quartets) and three atomic layers (triplets). Within each block the moments are aligned parallel and neighbouring blocks have their net moments aligned antiparallel. Thus a seven-layer structure may be considered as being made up of alternate quartets and triplets with the moments in the triplets pointing up and those in the quartets pointing down. The net result of this structure is that for each quartet–triplet pair there is the equivalent of the moment on one atomic layer pointing (in this arbitary case) down, hence a net ferromagnetic moment is produced. The formation of such a moment leads to a magnetostrictive deformation of the lattice and the competition between the elastic energy and the magnetoelastic energy will stabilize this structure. Gibbs *et al* identified three ferromagnetic structures which are stable over temperature ranges of up to 3.5 K. Those structures with an even ratio of quartets to triplets do not have a net ferromagnetic moment and do not appear to lock in. The possible combinations are presented in table 1 with the temperature limits of the stable phases. Each triplet may be thought of as corresponding to a spin-slip and hence the high symmetry structures may be characterized by a spin-slip distance, b, as used by Cowley and Bates [6]. This is the parameter that we will use for the remainder of the paper.

Table 1. Commensurate structures in the intermediate phase of Er, with their temperature limits as observed by Gibbs *et al* [1].

Propagation vector q	Spin-slip distance b	Repeat pattern	Ferro/ antiferro	Temperature range (K)
2/7	7	4,3	Ferro	51.5–48.5
3/11	11	4,4,3	Antiferro	
4/15	15	4,4,4,3	Ferro	34.5–32
5/19	19	4,4,4,4,3	Antiferro	
6/23	23	4,4,4,4,4,3	Ferro	26.5–32

Lin *et al* [7] have recently reported a detailed neutron scattering study of erbium in zero applied field and with a field applied along the c axis which reveals that the application of a c-axis field stabilizes commensurate structures in the intermediate region and is in broad agreement with our own measurements performed in an applied c-axis field [8]. In addition, they have shown that in the eight-layer structure and in the conical structure below T_c the basal plane anisotropy becomes important and that both these structures involve basal plane spin-slips. Their zero-field data shows a lock-in to the $b = 19$ structure from 34 K to 29 K, followed by a lock-in to the eight-layer structure, apparently in contradiction to the data of Gibbs *et al* which showed a lock-in to the $b = 15$ structure from 34.5 K to 32 K followed by a lock-in to the $b = 23$ structure from 26.5 K to 23 K.

The existence of commensurate structures at the temperatures proposed by Gibbs *et al* is supported by measurements of susceptibility [9], magnetization, and specific heat [10].

The single-crystal elastic moduli have been measured by several authors [11, 12]. The only measurements to show coupling which may be attributed to the formation of high-symmetry phases are those of Jiles and Palmer [12], who observed anomalous behaviour of the elastic moduli at 26.5 K, 33 K and 42 K. The ultrasound measurements of Ho by Bates *et al* [13], demonstrated the suitability of such measurements for delineating and identifying commensurate phases in spin-slip structures.

We have measured the ultrasonic elastic moduli C_{33}, C_{44}, C_{11} and C_{66} and their associated attenuation coefficients, α, for a single crystal of erbium with a view to correlating our data with the existing spin-slip models for the magnetic structure.

2. Experimental details

The single crystal of erbium was grown by solid state methods at the School of Metallurgy and Materials, University of Birmingham, by Dr D Fort, and is believed to have a purity of 99.99%.

All the measurements were made using 15 MHz quartz transducers bonded to the sample by epoxy resin in conjunction with an automatic computer controlled system described elsewhere [14] which has been shown to provide excellent point to point sensitivity, but is susceptible to absolute errors of up to 3% in the elastic constant. The attenuation coefficient was calculated from the ratio of the amplitudes of two successive echoes. No attempt has been made to obtain absolute values of attenuation by making appropriate corrections.

The temperature was controlled using a conventional continuous flow cryostat and controller, allowing measurements to be made down to 10 K with an accuracy of ±0.2 K.

3. Experimental results

3.1. C_{33} and α_{33}

The C_{33} elastic modulus is derived from the velocity of a longitudinal wave propagating along the *c*-axis. All previous measurements of C_{33} show dramatic coupling to the three magnetic phase transitions. Initial measurements performed at a cooling rate of 1 K min^{-1} show the sharp step-like features coresponding to the Néel point and the onset of a basal plane moment at 52 K (T_m) (figure 1); however, the nature of the transition at T_c ($\simeq$ 18 K) differs quite considerably from that observed by other authors.

As the temperature is reduced C_{33} appears to undergo a hardening starting at approximately 28 K with a pronounced inflection at 23 K. The hardening continues until the rapid drop of 5% at 18 K corresponding to the onset of the conical structure. In comparison, Jiles and Palmer [12], and DuPlessis [11], observed reductions of approximately 4% and 2.5% respectively. In addition, both sets of data show some softening for approximately 3 K above T_c.

A more thorough investigation of the intermediate phase at a cooling rate of approximately 0.5 K min^{-1} (figure 1 (inset)) showed a plateau-like feature between 49.5 K and 47.5 K and a modest dip at 33 K.

The attenuation coefficient, α_{33}, couples strongly to the three principal phase transitions, but showed no anomalous behaviour which could be associated with the formation of commensurate phases.

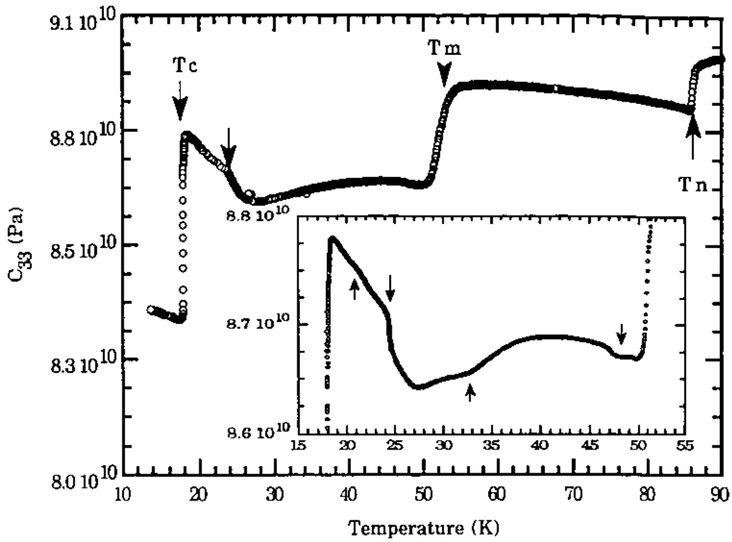

Figure 1. The temperature dependence of C_{33} cooled at 1 K $\min^{-1}$ and 0.5 K $\min^{-1}$ (inset).

3.2. C_{44} and α_{44}

The C_{44} elastic modulus may be derived from the velocity of a shear wave polarized parallel to the c axis propagated across the basal plane, or the velocity of a shear wave propagated parallel to the c axis with any polarization within the basal plane. It has long been reported [15] that in the presence of a magnetic field, or in a ferromagnetic phase, the degeneracy of these two modes would be lifted. This effect is apparent in the measurements of C_{44} by duPlessis [11], and becomes more pronounced with increasing applied magnetic field. Both modes have been measured (figure 2), and a difference between the two measurements of both α_{44} and C_{44} becomes obvious below T_m. In both cases the signal becomes immeasurably small below 21 K. C_{44} shows inflections at 24 K and a smaller feature at 27 K for both modes. The differences between the two modes are most clearly visible in the α_{44} data. In both cases a peak is visible at 24 K prior to the dramatic and catastrophic increase in the attenuation. The peak is much more pronounced on the c-axis mode than on the a-axis mode and is clearly resolved from the increase in the attenuation which results in the loss of the signal which appears to peak at approximately 21 K. Both modes exhibit a broad feature between 27 K and 30 K.

Measurements between 21 K and 18 K proved to be impossible with decreasing temperature. However, measurements could be made on warming the sample up from 15 K. At T_c, C_{44} experiences a dramatic softening accompanied by a rapid increase in the attenuation, and ultimately the disappearance of the signal (inset to figure 2).

3.3. C_{11}, α_{11}, C_{66} and α_{66}

The C_{11} elastic constant and the α_{11} attenuation coefficient correspond to a longitudinal wave propagated parallel to the basal plane. In the intermediate region,

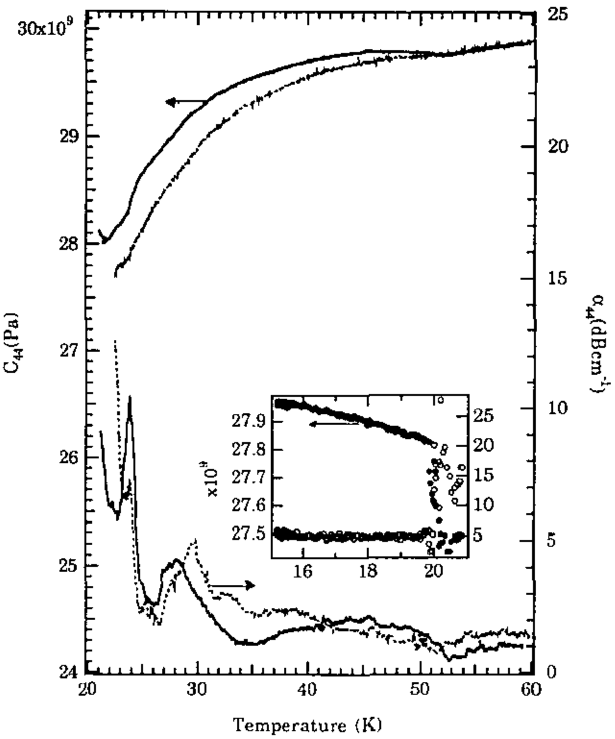

Figure 2. The temperature dependence of C_{44} and α_{44} between 60 K and 20 K and (inset) between 15 K and 21 K. The dashed lines correspond to a shear wave polarized parallel to the *c* axis propagated across the basal plane.

C_{11} experiences considerable attenuation and the signal was immeasurably small between 47 K and 27 K.

On cooling from 59 K through T_m the elastic constant is seen to soften from 53 K (figure 3), with the softening becoming less steep between 51 K and 49 K. The softening of C_{11} is accompanied by an increase in α_{11}, measured over the same temperature range. Two modest peaks can be seen at 48.5 K and 50 K.

The data collected while warming the sample from 15 K to 27 K (figure 4) show more structure. A dramatic hardening in C_{11} at 20 K heralds the phase transition, above which C_{11} softens gradually showing a minimum at 26.5 K. The α_{11} data display shows strong coupling to the transition at 20 K and a very distinct feature centred on 26.5 K coincident with the minimum in C_{11}.

C_{66} and α_{66}, corresponding to a shear wave propagating parallel to the basal plane and polarized in the basal plane, have also been measured. High attenuation prohibited measurements between 20 K and 51 K, consequently no coupling to commensurable structures was observed. Both α_{66} and C_{66} coupled to T_c and T_m.

4. Discussion

The C_{33} data reveal quite distinct effects at 48 K and 33 K. By correlation with the data

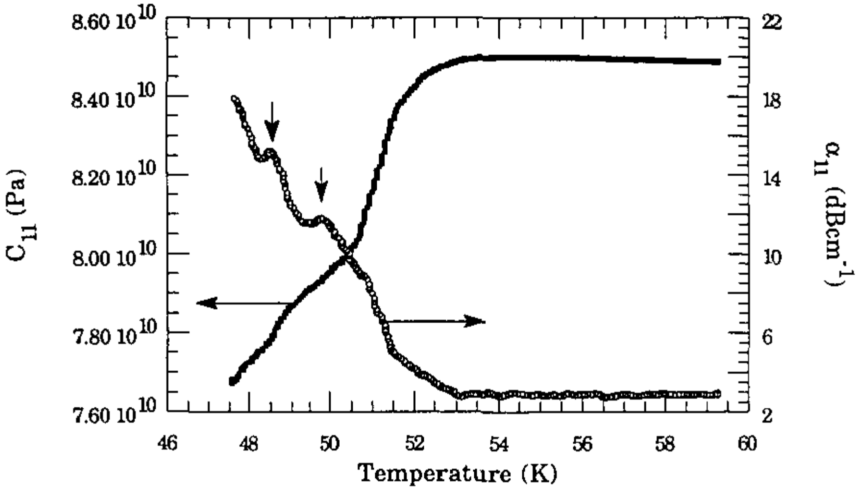

Figure 3. The temperature dependence of C_{11} and α_{11} between 60 K and 46 K.

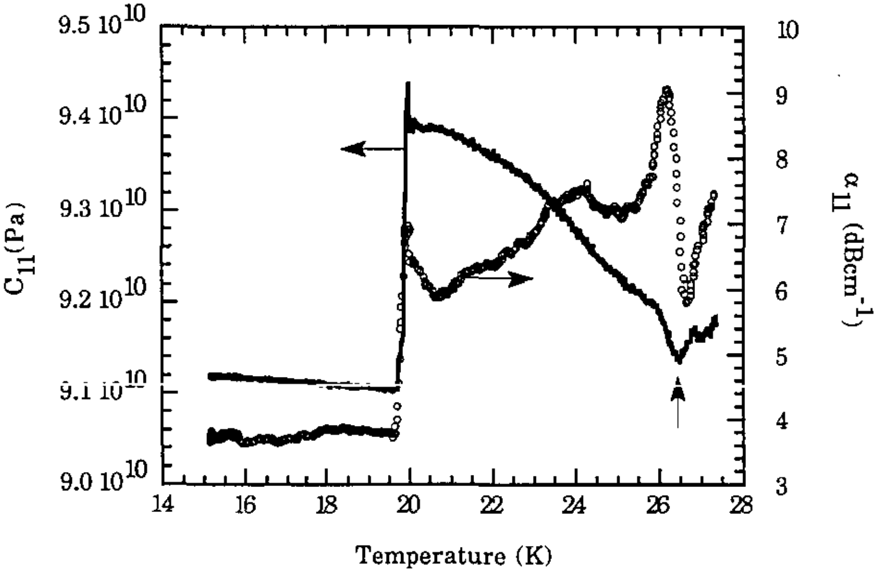

Figure 4. The temperature dependence of C_{11} and α_{11} between 15 K and 28 K.

of Gibbs *et al* [5], these would seem to be the result of the formation of the $b = 7$ and $b = 15$ structures respectively; on the other hand the data of Lin *et al* [7] would suggest that the anomaly at 33 K was derived from the formation of the $b = 19$ phase, which does not have a net ferromagnetic moment. The broad nature of both anomalies may be the result of locking in to the respective commensurate structures. It is interesting that these anomalies were only visible when the sample was cooled or warmed very slowly. At 24 K a sharp knee in C_{33} was visible, irrespective of the cooling rate, and would seem to correspond to the low temperature limit of the $b = 23$ structure.

The C_{44} data show a very small inflection at approximately 27 K, which is accompanied by a broad increase in α_{44} over the region 32 K to 27 K. The shape of this feature is not repeatable and its origins are not well understood although it may arise as a result of the formation of the $b = 15$ structure.

At 24 K α_{44} displays a pronounced peak which is accompanied by a small change in C_{44}. Our own neutron scattering data [16] collected for the same crystal suggest that the peak is coincident with the lower temperature limit of the $b = 23$ phase. This behaviour is however reminiscent of the peak in α_{44} observed by Lee *et al* [17] at 95.5 K in Ho, where the small anomaly in C_{44} at the same temperature has been attributed to the formation of a $b = 2$ spin-slip structure in the basal plane [13]. However it is not clear why this structure produces such a dramatic peak in the attenuation. The increase in α_{44} below 24 K, which causes the signal to disappear at 21 K, may arise as a result of the formation of the eight-layer structure. Lin *et al* [7] have shown that as the temperature decreases, the importance of the basal plane anisotropy increases and the spins adopt spin-slip structures in the basal plane in the eight-layer structure. The rotation of the spins out of the *a*–*c* plane, to form something akin to an alternating cone structure, may produce the dramatic softening of C_{44}.

The C_{11} and α_{11} data provide evidence of a lock-in to the $b = 7$ structure between 50 K and 48.5 K, in good agreement with the C_{33} data and the data of Gibbs *et al.* The data collected while warming from the low-temperature phase show a pronounced feature in both C_{11} and α_{11} centred on 26.5 K. Taking the hysteresis into account this feature may be associated with the formation of the $b = 23$ phase.

The ultrasound data described above are summarised in table 2 and the spin-slip parameter of the structure which is likely to have produced the anomalous behaviour is given. The difference in the data collected by Lin *et al* and those collected by Gibbs *et al* in addition to discrepancies between other authors suggest that there is some sample dependence on the tendency of the magnetic structures to adopt particular commensurate structures.

Table 2. Temperature of observed anomalies. Where appropriate, the b values of the structures which are thought to give rise to the anomalies are given. All data were collected with decreasing temperature unless indicated otherwise by a w.

	T_c (K)		$b = 23$	$b = 15$	$b = 7$	T_m (K)	T_N (K)
C_{33}	18/20w		24	33	47.5–49.5	52	85
α_{33}	18					52	85
C_{44}	20w	21.5/22	24			52	85
α_{44}	20w	21.5/22	24–27				
C_{11}	20w		26–28w		48	52	85
α_{11}	20w		26–28w		48.5–49.5	52	
C_{66}	20w					53	85

The coupling of commensurate structures to the elastic constants is expected to be dominated by the magnetostatic deformation of the lattice on the formation of the ferrimagnetic structures. Commensurate structures without a net ferromagnetic moment may couple to the elastic constants as a result of symmetry breaking or domain relaxation effects [13], however, no such coupling has been observed in this study. This would seem to be in agreement with measurements of other properties, which in general show large effects in the ferrimagnetic regions and only small effects, if any, in the antiferromagnetic regions.

5. Conclusion

We have measured the elastic constants C_{33}, C_{44}, C_{11} and C_{66} and their associated attenuation coefficients. All bar C_{66} show anomalous behaviour in the region 53 K to 18 K which we attribute to the formation of ferrimagnetic commensurate structures in accordance with the spin-slip model of Gibbs *et al.*

The disproportionately large coupling of α_{44} to the $b = 23$ structure, in comparison to other commensurate structures, and the high value of α_{44} in the eight-layer region may correspond to the growing importance of the basal plane component of the magnetization as the temperature decreases. Further ultrasound measurements performed with a magnetic field applied parallel to the c axis and in the basal plane may yield more information about the orientation of the spins in this region.

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