ULTRASONIC STUDY OF ANTIFERROMAGNETIC DOMAINS IN DYSPROSIUM

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ABSTRACT — Measurements of thermal hysteresis in the elastic constant C_{33} and its associated ultrasonic attenuation α_{33} for single crystal dysprosium have been carried out using CdS thin film transducers. The results are very similar to earlier ones using quartz transducers. bonded whith epoxy resin. The anomalous behaviour is interpreted in terms of antiferromagnetic spiral spin domains and this view is supported by thermal cycling studies.

INTRODUCTION

The heavy rare earth metals display a wide range of magnetic spin structures, one of the most fascinating being the antiferromagnetic spiral spin or helical structure. The magnetic moments are confined to the basal planes of the hexagonal structure and in a particular plane all the moments are ferromagnetically aligned. As one moves along the hexagonal c-axis, however, the moments rotate from basal plane to basal plane by a constant angle, θ, the turn angle. Among the elements, Tb, Dy and Ho have a spiral spin state as do the intra-rare earth alloys Tb-Ho and Gd-Y. The antiferromagnetic order commences at the Néel Temperature T_N and disappears at the Curie Temperature T_c. Values of T_N and T_c for the elements and for selected alloys are shown in Table I.

TABLE I

Material	T_N	T_c
Tb	226 K	219 K
Dy	180 K	~ 85 K
Ho	139 K	20 K
Tb-50 % Ho	185 K	~ 85 K
Gd-40 % Y	183 K	~ 95 K

The spiral spin structure has attracted a great deal of interest in recent years and it was soon realised that a single crystal sample containing a continuous spiral of magnetic moments would act as a polarizer of an incident neutron flux. However, no such preferential

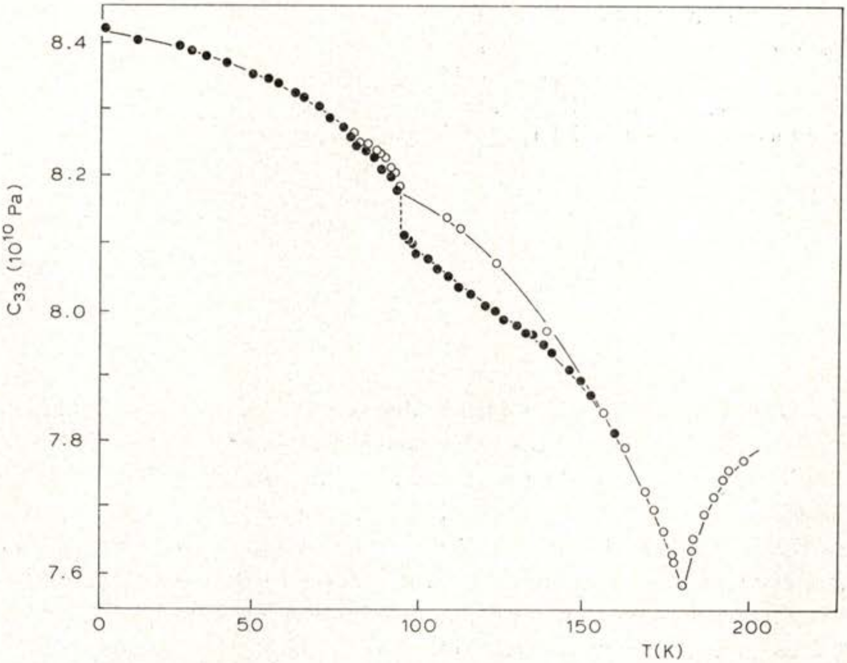

Fig. 1 — Temperature dependence of the elastic constant C_{33} (in units of 10^{10}Pa) for Dysprosium [using quartz transducers]

○ Temperature decreasing
● Temperature increasing

polarization was observed and it was therefore assumed that a particular sample is divided into regions where the spiral twists either clockwise or anticlockwise with respect to the c-axis.

Further evidence for the presence of these two types of region or domain was provided by studies of the ultrasonic propagation in the heavy rare earths. The elastic constant C_{33}, measured by propagating compressional waves down the hexagonal axis, and its associated ultrasonic attenuation α_{33} display marked thermal hysteresis in the spiral spin regime of Dy, Tb-50% Ho and Gd-40% Y [1]. The measurements reported in [1] show an anomalous drop in C_{33} and a sharp rise in α_{33} when the sample is warmed from below T_c. Figures 1 and 2 show the results obtained for Dy. The ultrasound was generated and detected with quartz transducers which were bonded to the sample with Araldite epoxy resin. The epoxy resin bond was neces-

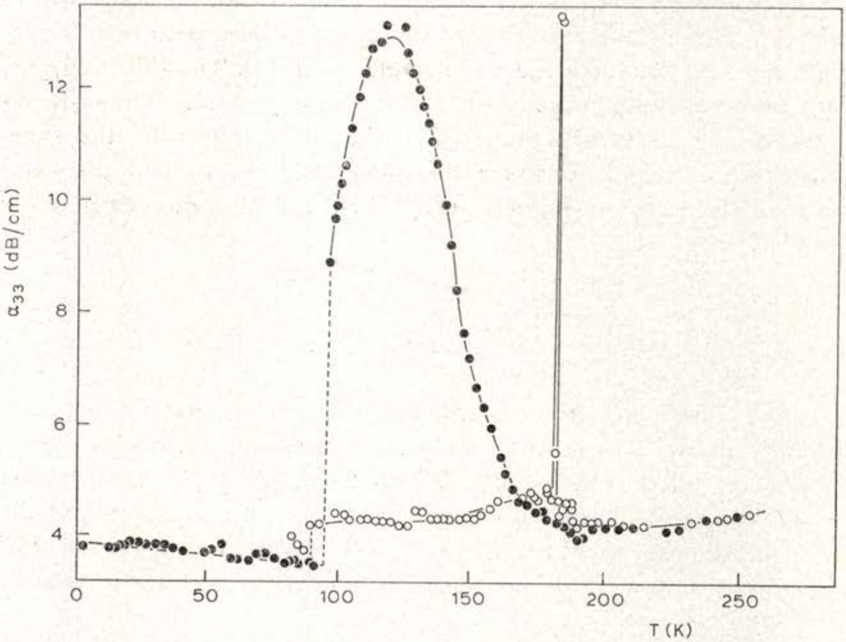

Fig. 2 — Temperature dependence of the ultrasonic attenuation α_{33} (db/cm) for Dysprosium [using quartz transducers]

○ Temperature decreasing
● Temperature increasing

sary because the large differential thermal expansion between the rare earth sample and the quartz transducer was sufficient to break weaker bonding agents.

It is possible, however, that the use of quartz transducers and Araldite bonds will clamp the relatively soft rare earth sample preventing it from developing its full strain. This clamping could affect the c-axis lattice parameter and hence the inter-layer turn angle of the magnetic spiral. To check the reliability of the measuring technique we have replaced the quartz transducers with CdS thin film transducers [2]. These have been shown to adhere to rare earth metals over wide ranges of temperature and sample strain [3]. The thin film puts very little constraint on the sample and simply breaks into «island» areas at high states of strain. Dysprosium was chosen for the experiment since it had a wide antiferromagnetic region and the sample available was of higher purity than the intra-rare earth alloy samples.

EXPERIMENTAL DETAILS AND RESULTS

The elastic constant C_{33} was calculated from measurements of the velocity of longitudinal waves, propagating down the c-axis, using a standard pulse echo overlap technique [5,6]. Temperature variation was achieved with a standard gas flow cryostat and the temperature was measured by a 0.03 % Fe in Au versus chromel thermocouple to an absolute accuracy of ± 0.5 K, while the relative accuracy was ± 0.1 K.

The single crystal of Dy was in the form of a cylinder, 5 mm in diameter and 35 mm long, cut with its faces perpendicular to the c-axis. It was grown by zone flotation at Metals Research Ltd. and had a purity of better than 99 atomic %. The CdS transducers were vacuum evaporated onto the surface of the sample with a sufficient thickness to allow operation at 15 MHz. Since this was the resonant frequency of the quartz transducers it allowed direct comparison with the earlier measurements [1].

The measurements of C_{33} and α_{33} for Dy taken while the temperature was reduced to 4.2 K and while it was increased from 4.2 K, i.e., from the ferromagnetic region, to room temperature are shown in Figs. 3 and 4. The results obtained for C_{33} are very similar to the previous measurements and exhibit a nearly identical hysteresis when

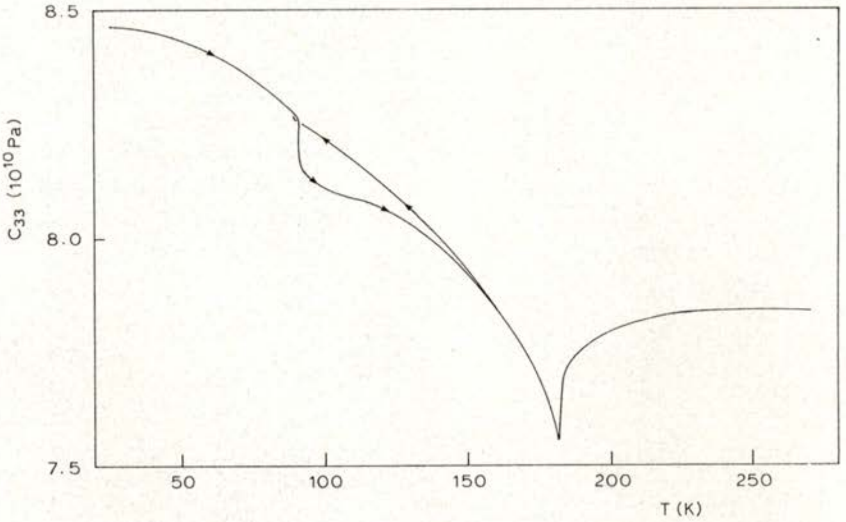

Fig. 3 — Temperature dependence of the elastic constant C_{33} (in units of 10^{10}Pa) for Dysprosium, [using CdS transducers]. Arrows indicate direction of temperature change.

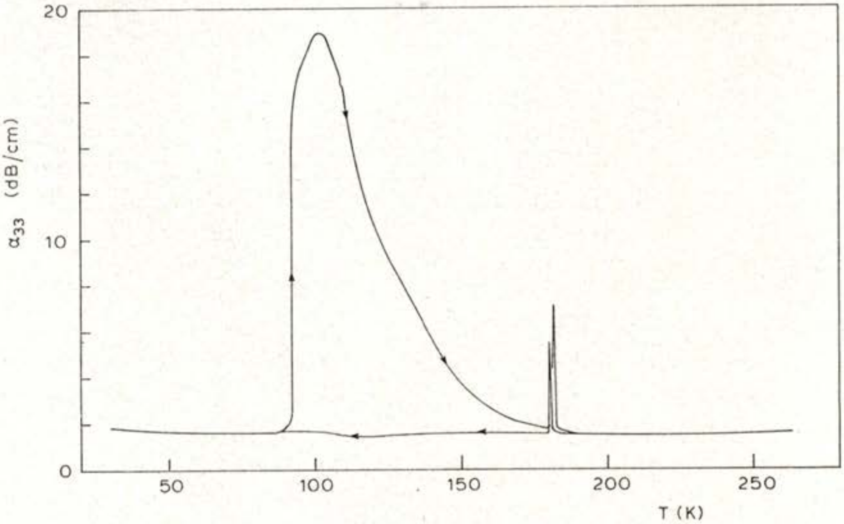

Fig. 4 — Temperature dependence of the ultrasonic attenuation α_{33} (db/cm) for Dysprosium [using CdS transducers]. Arrows indicate direction of temperature change.

warming from below T_c. The results for α_{33} are again similar except that the phase change at T_c is not seen when cooling and the anomalous attenuation peak is much higher with CdS transducers.

Figure 5 shows the ultrasonic attenuation close to T_N. The hysteresis of 1.5K in T_N is of interest since it is well outside the experimental error associated with thermal equilibrium of the sample. However, more important in the present context is the presence of thermal hysteresis in α_{33} below T_N which tends to zero as T_N is approached. It should be noted (Fig. 6) that the thermal hysteresis in C_{33} does not extend above 160K.

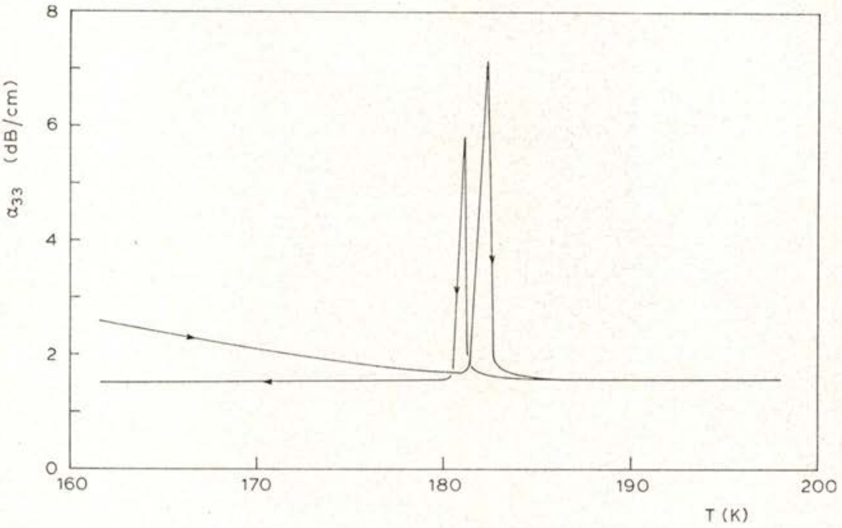

Fig. 5 — Temperature dependence of the ultrasonic attenuation α_{33} (db/cm) for Dysprosium in the vicinity of T_N. The arrows indicate the direction of temperature change.

Figure 7 shows in more detail the sharp rise in α_{33} close to T_c associated with the onset of the spiral spin state. Figure 8 (a) shows the effect of thermal cycling in the region of T_c. The sample is first cooled below T_c into the ferromagnetic phase and then warmed into the spiral spin phase where C_{33} drops as expected. The warming of the sample was then halted at three separate temperatures, 100K, 109K and 123K and the sample cooled below T_c while the behaviour of C_{33} was monitored (Fig. 8 (a, b)).

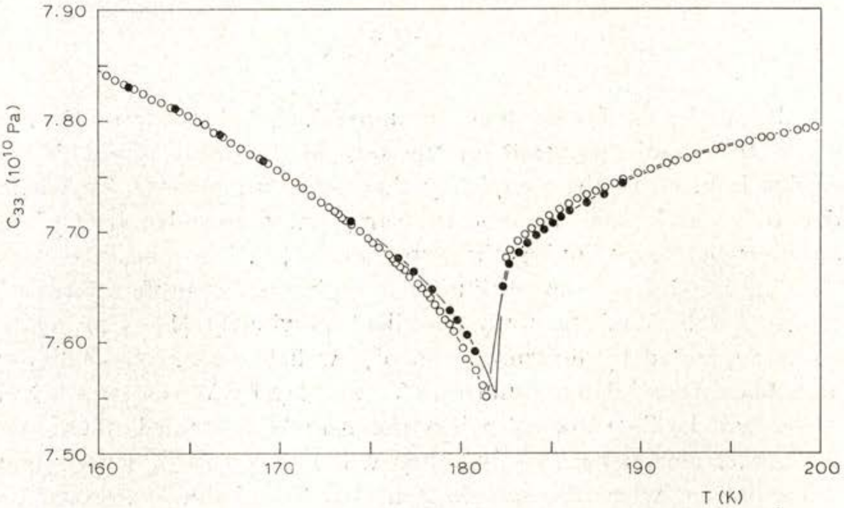

Fig. 6 — Temperature dependence of the elastic constant C_{33} (10^{10}Pa) for Dysprosium in the vicinity of T_N.

- o Temperature decreasing
- • Temperature increasing

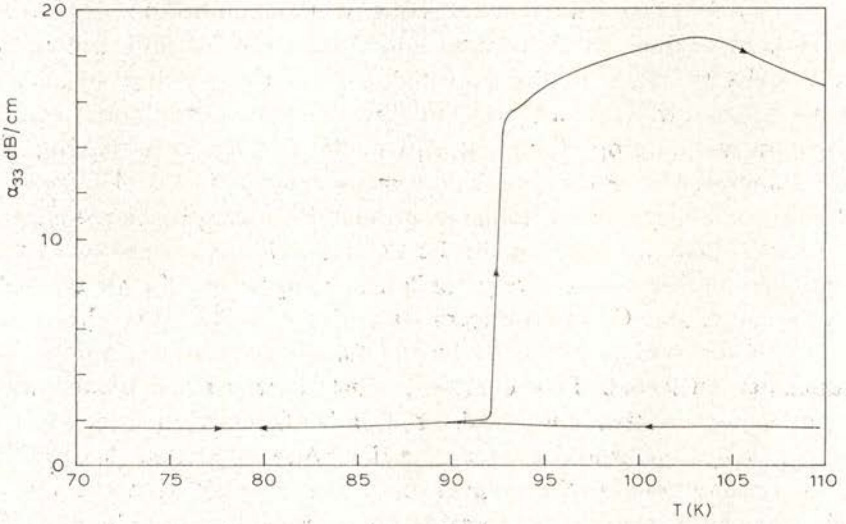

Fig. 7 — Temperature dependence of the ultrasonic attenuation α_{33} (db/cm) for Dysprosium in the vicinity of T_c.

DISCUSSION

It can be confirmed that the anomalous behaviour of C_{33} and α_{33} in the spiral spin state of the rare earth metals is not due to strains induced in the sample by the measuring process. To ensure that this was indeed the case the sample was annealed at 60° C for an hour before beginning the experiments [7]. The rise in α_{33} and the fall in C_{33} can be attributed to spiral spin antiferromagnetic domains (SSD) of the form described previously [1]. This analysis is supported by measurements of low field (a.c.) susceptibility and thermal modulation studies in Tb and Dy by Wilson et al [8, 9]. There will be SSD present when the sample is cooled down into the antiferromagnetic state but they will be far less numerous than those present when the sample is heated from below T_c. Extra SSD will be nucleated from the ferromagnetic Bloch walls in a manner described earlier [1]. The domain boundaries between SSD will be ferromagnetic and the lowering of the elastic constant C_{33} will be caused by the well known «Δ E» effect. The applied stress of the acoustic wave produces rotation of the magnetisation in the domain wall developing a magnetostrictive strain that adds to the elastic strain reducing the elastic constant below its non-magnetic value.

The ferromagnetic domain walls will be pinned to impurities and imperfections in the crystal and the ultrasonic wave will cause the walls to move in the potential well of the pinning site. This phenomenon will obviously lead to ultrasonic attenuation and the attenuation will be proportional, at a particular frequency, to the number of domains. Hence the attenuation measurements in Fig. 4 infer that there are always more domains present in a sample warmed from below T_c than would normally be the case, although close to T_N the numbers of domains are very similar. A comparison of Figs. 2 and 4, with quartz and CdS transducers respectively, indicate that the clamping of the sample produced by the former reduces the number of domains nucleated. This comment must, however, be treated with caution as the attenuation peak itself varies from run to run.

Finally one would expect from the «ΔE» effect that the drop in C_{33} is proportional to the number of domains present. Hence in Fig. 8 the maximum number of domains are nucleated close to T_c and this number decreases as T_N is approached. However, when the temperature is lowered from an intermediate value the number of domains

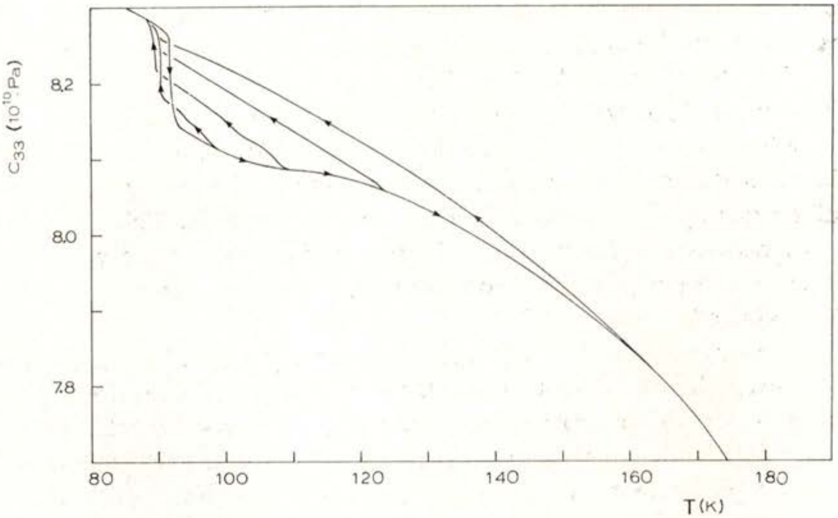

Fig. 8(a) — Temperature dependence of the elastic constant C_{33} (10^{10}Pa) for Dysprosium in the vicinity of T_c .

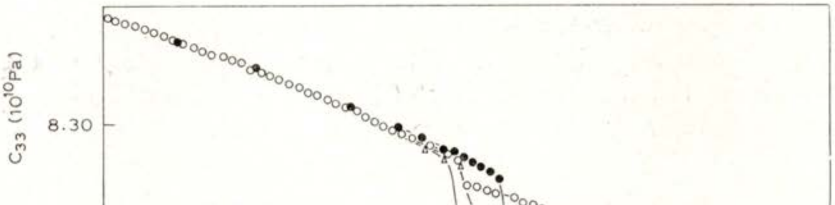

Fig. 8(b) — Enlarged version of 8(a) in the region of T_c .

- ○ Temperature decreasing from 300 K.
- ● Temperature increasing from 4 K.

Temperature increase from below T_c followed by:

- △ Temperature decreasing from 109 K.
- ▲ Temperature decreasing from 123 K.

remains essentially constant, with all four curves being very nearly parallel, until the Curie temperature is reached.

We have available, therefore, a technique for increasing the number of SSD, whereas the requirement for a neutron polarizer is to reduce the number of domains present. It is probable that fewer domains can only be produced by improving the purity and crystalline quality of the single crystal samples available. This improvement will reduce the number of pinning sites for domain walls and hence the number of domains, especially when the sample is cooled from above T_N.

The hysteresis in T_N, (Figs. 5,6) probably arises from the different strain levels present in the Dy crystal when cooling from room temperature and warming from below 100 K. It was reported previously [7] that the ordering temperatures of the rare earth metals are strongly dependent on the state of strain in the sample. It should be noted that the measurement of ultrasonic attenuation associated with spin fluctuation scattering at the phase change is a remarkably sensitive tecnhique for defining the transition temperature and therefore studying any hysteresis.

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