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# Temperature modulation of the antiferromagnetic susceptibility of high-purity single-crystal terbium

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**Abstract.** Detailed measurements of the AC magnetic susceptibility $\chi'(T)$ and its response $S(T) \equiv \Delta\chi'(T)/\Delta T$ to a thermal modulation wave have been carried out around the narrow helical antiferromagnetic (AF) region (about 221–229 K) of a high-purity single crystal of terbium, along its magnetically easy $b$ axis. We have extended existing theories of AF susceptibility of spiral spin structures to demonstrate that the number of spins in the intervening domain wall should decrease with increasing temperature. The $\chi'(T)$ and $S(T)$ results obtained on cooling into, and warming from, the AF region, without crossing $T_C$, are consistent with this theory. Significantly enhanced AF susceptibility and corresponding effects in $S(T)$ are observed on warming from the ferromagnetic region into the AF region. This behaviour can be accounted for by a spatial alignment of ferromagnetic islands centred on magnetic inclusions and/or thicker aligned domain walls of ferromagnetic character that separate AF domains of opposite chirality.

## 1. Introduction

In this paper we present the findings of a detailed AC magnetic susceptibility study of the antiferromagnetic (AF) region of a high-purity single crystal of terbium. The results and analyses are based primarily on the temperature modulation technique in which the response of the in-phase AC magnetic susceptibility $\chi'$ to a thermally modulated wave $\Delta T$ is determined by $S(T) \equiv \Delta\chi'(T)/\Delta T$. Previously we have presented a temperature modulation study of the helical spin AF region of polycrystalline dysprosium (McKenna *et al* 1983). The results were consistent with the existence of spiral spin (SS) domains in the AF region (Palmer 1975) and with the formation of an SS domain structure which was found to be different on warming the sample from the ferromagnetic region to the AF region, compared with that obtained when the sample was cooled from the paramagnetic region to the AF region.

The qualitative behaviour of the magnetic susceptibility of dysprosium in the AF region has been discussed by del Moral and Lee (1974) in terms of the magnetic response of spins with an SS structure. They also suggested that quantitative disagreements between theory and experiment might be explained by an additional contribution due to the presence of 'ferromagnetic' domain boundaries separating AF domains. The contributions to the susceptibility can be expressed as (McKenna *et al* 1983)

$$\chi'(T) = \chi_D(T) + \chi_W(T) \tag{1}$$

where $\chi'(T)$ is the measured in-phase component of the AC magnetic susceptibility, $\chi_D(T)$ is the response of spins within the spiral structure, i.e. within the SS domains (del Moral and Lee 1974, Kitano and Nagayima 1964), and $\chi_W(T)$ is the contribution due to the domain walls which separate regions having an opposite sense of spin spiral or chirality. The experimentally observed hysteresis in $\chi'(T)$ for polycrystalline impure dysprosium in the AF region (McKenna *et al* 1983) was attributed to hysteresis in the component $\chi_W(T)$ of equation (1) with the warming value being greater than the cooling value, i.e. $\chi_W(T)_\uparrow > \chi_W(T)_\downarrow$. Hysteresis in this domain wall contribution to $\chi'(T)$ should therefore be due to some irreversible change in the domain wall structure as the temperature is varied. It was clear that the degree of hysteresis was fundamentally dependent on whether or not warming runs embraced a transition of $T_C$, but the broadness of the transition prevented a detailed study.

Terbium has a similar helical spin structure to that of dysprosium but over a more restricted AF region (about 221–229 K for terbium compared with about 90–180 K for dysprosium). Neutron topographic studies of terbium revealed domain structures in the AF region which differ for warming and cooling (Baruchel *et al* 1981, Palmer *et al* 1986). In addition, previous studies of the AC magnetic susceptibility of single-crystal terbium and its transient enhancement (TE) have also yielded results consistent with the existence of such domains (McKenna *et al* 1981a). The same high-purity terbium single crystal used in the present investigation allows delineation of the transition from the ferromagnetic region to the AF region to within 0.5 K. By comparison, the rounding at $T_C$ in the less pure polycrystalline dysprosium sample (McKenna *et al* 1983) was about 20 K or less. In this paper the behaviour of the high-resolution thermal modulation results for terbium, as well as the earlier results for dysprosium, is discussed in terms of a simple theory of SS domains and compared with the neutron topographic studies of Baruchel *et al* (1981), Baruchel *et al* (1986) and Palmer *et al* (1986).

## 2. Theory: domain walls in the helical antiferromagnetic region

It should be noted that the following theory pertains solely to the AF region and does not include temperature-related memory effects which may occur as a result of crossing phase transitions. As the nature of $\chi_W(T)$ (equation (1)) in the AF region will be influenced by the number $n_W$ of spins in a domain wall, it was decided to extend the earlier theoretical work of Thomas and Wolf (1965) in order to estimate $n_W$ and in particular to examine the temperature dependence of $n_W$. For brevity we henceforth refer to domain walls in the helical AF region as AF domain walls in order to distinguish them from domain walls in the ferromagnetic phase.

Thomas and Wolf (1965) calculated $E_W$, the energy of AF domain walls, using the general form of the three-plane interaction model in which the exchange energy $E_{ex}/N$ per atom in the spiral structure is expressed as

$$E_{ex}/N = -\sigma^2[B_0 + 2B_1\cos(\theta_0) + 2B_2\cos(2\theta_0)] \tag{2}$$

where $\sigma$ is the reduced magnetization, $\theta_0$ is the spiral turn angle and $B_0$, $B_1$ and $B_2$ are exchange constants (Elliott 1961). Using this model a stable structure exists for the conditions

$$B_2 < 0 \tag{3a}$$

and

$$\cos(\theta_0) = B_1/4|B_2|. \tag{3b}$$

The energy $E_W$ of an AF domain wall can be written as

$$E_W = E_{WS} - n_W E_{ex}/N \tag{4}$$

where $E_{WS}$ is the energy of the $n_W$ spins in a wall and $n_W E_{ex}/N$ is the energy which these spins would have in a spiral structure. Using equations (2) and (3*b*) and dropping the $B_0$ term which is common to both $E_{ex}/N$ and $E_{WS}$, we obtain

$$(E_{ex}/N)/2|B_2|\sigma^2 = -[2\cos^2(\theta_0) + 1]. \tag{5}$$

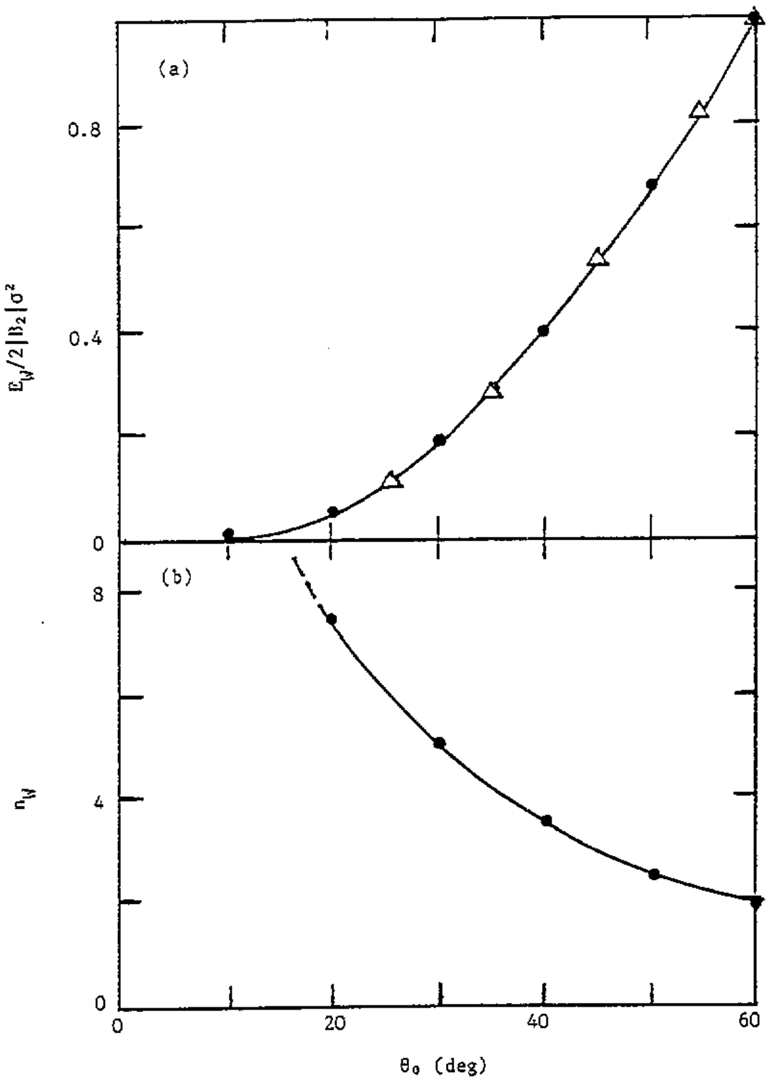


**Figure 1.** (*a*) The minimum value of the domain wall energy $E_W/2|B_2|\sigma^2$, plotted as a function of turn angle $\theta_0$. The present calculations (●) are compared with the values derived from the earlier result of Thomas and Wolf (1965) (△). (*b*) A plot of $n_W$ for the minimum energy of an AF domain wall as a function of $\theta_0$ ($n_W$ is the number of spins in the wall).

We assume that there is a constant change in turn angle per spin through the wall, varying from $+\theta_0$ for the initial spin to $-\theta_0$ for the $n_W$ spin. Again dropping the $B_0$ term and using equations (2) and (3$b$), the energy $E_{WS}$ for spin $i$ in a wall can be represented by

$$E_{WS}/2|B_2|\sigma^2 = -\{2\cos(\theta_0)[\cos(\theta_i) + \cos(\theta_{i-1})] - \tfrac{1}{2}[\cos(\theta_i + \theta_{i+1}) + \cos(\theta_{i-1} + \theta_{i-2})]\}. \quad (6)$$

$E_W/2|B_2|\sigma^2$, the normalized energy of a wall containing $n_W$ spins, is determined by subtracting equation (5) from equation (6) for the spin assembly comprising the $n_W$ spins of the wall and the nearest-neighbour spins at each end of the wall. For any value of $\theta_0$, the most stable AF domain wall should contain a number $n_W$ of spins for which the value $E_W/2|B_2|\sigma^2$ is a minimum. Figure 1($a$) shows these values of $E_W/2|B_2|\sigma^2$ plotted as a function of $\theta_0$ for several values of $\theta_0$ from 10° to 60°. The good agreement with values taken from figure 5 of Thomas and Wolf (1965) suggests that the above procedure is equivalent to their numerical iteration method of calculation. Using the present procedure it is also possible to produce an analytical solution for the condition $\theta_0 = 60°$, giving $E_W/2|B_2|\sigma^2 = 1$ and $n_W = 2$, in agreement with the results of Thomas and Wolf (1964).

Recently Fairbairn and Singh (1986) used a continuum approximation to calculate domain wall widths for a fixed $\theta_0$, allowing a continuous variation in turn angle through the wall. When using the three-plane interaction model for $\theta_0 = 37°$ they report an $n_W$ of five lattice spacings—a result similar to our value of $n_W = 4$ for the same $\theta_0$ (figure 1($b$)). We are therefore confident in extending the simple model used by Thomas and Wolf (1965) to examine the temperature dependence of $n_W$.

Figure 1($b$) is a plot of $n_W$ for minimum $E_W$ as a function of turn angle $\theta_0$. The results of this analysis indicate that the number of spins in the most stable wall decrease with increasing $\theta_0$. Thus, for terbium and dysprosium where $\theta_0$ generally increases with increasing temperature (Koehler 1972), the calculations presented here suggest that $n_W$ should decrease with increasing temperature.

The temperature dependence of the domain wall energy $E_W$ can be calculated for a particular materal with knowledge of the magnetization and temperature dependence of $B_2$. For dysprosium, Elliott (1961) determined $B_2$ (in kelvins) as

$$|B_2| = (57 - 11\sigma^2). \quad (7)$$

Using the experimentally determined values for dysprosium (Koehler 1965) (the relevant data are reproduced in figure 2($a$)), it is possible to calculate $n_W$ for the minimum value of $E_W$ as a function of temperature (figure 2($b$)). $E_W$ can also be determined (figure 2($c$)) from the theoretical dependence of the relative magnetization $\sigma(T)$ (Morrish 1965). (The equivalent representation of $B_2$ from equation (7) for terbium was not available.)

If it is assumed that the contribution of AF domain walls to the susceptibility $\chi_W(T)$ is dependent on $n_W(T)$, then hysteresis in $\chi_W(T)$ could be accounted for by hysteresis such that $n_W(T)$ on warming is greater than $n_W(T)$ on cooling, i.e. $n_W(T)_\uparrow > n_W(T)_\downarrow$. This model explains the hysteresis observed in $\chi'(T)$ for dysprosium when the sample is cooled from the paramagnetic region and cycled within the AF region. However, the behaviour of the thermal modulation response $S$ for dysprosium when warming from below $T_C$ (figure 1($c$) of McKenna *et al* (1983)) is not explained completely by this hysteresis model. Rather, it was found that the temperature modulation signal $S_\uparrow$ on warming showed an opposite sign to that of $(\mathrm{d}\chi'/\mathrm{d}T)_\uparrow$ (the analytical derivative of the

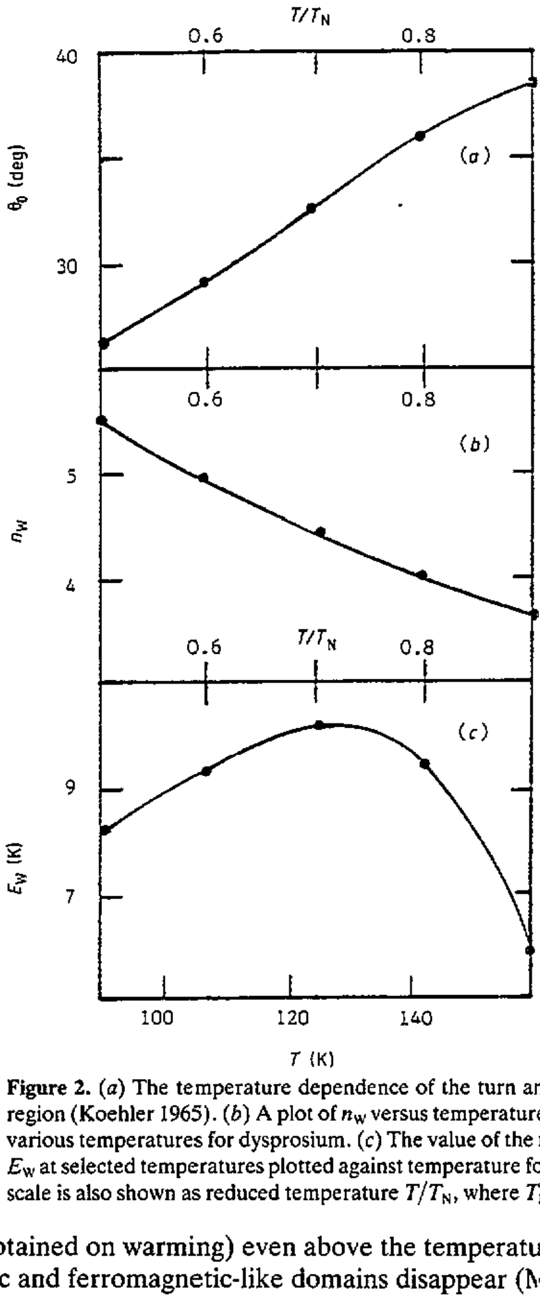


**Figure 2.** (*a*) The temperature dependence of the turn angle $\theta_0$ for dysprosium in the AF region (Koehler 1965). (*b*) A plot of $n_W$ versus temperature for the minimum value of $E_W$ at various temperatures for dysprosium. (*c*) The value of the minimum AF domain wall energy $E_W$ at selected temperatures plotted against temperature for dysprosium. (The temperature scale is also shown as reduced temperature $T/T_N$, where $T_N$ is the Néel temperature).

$\chi'(T)$ data obtained on warming) even above the temperature (about 120 K) at which ferromagnetic and ferromagnetic-like domains disappear (McKenna *et al* 1980). With the benefit of the far sharper transition observed in the present study of the high-purity single-crystal terbium (section 4), the above behaviour of $S_\uparrow$ for dysprosium together with the corresponding behaviour for terbium is explained in section 5.

## 3. Experiment

Details on the AC magnetic susceptibility and temperature modulation techniques have

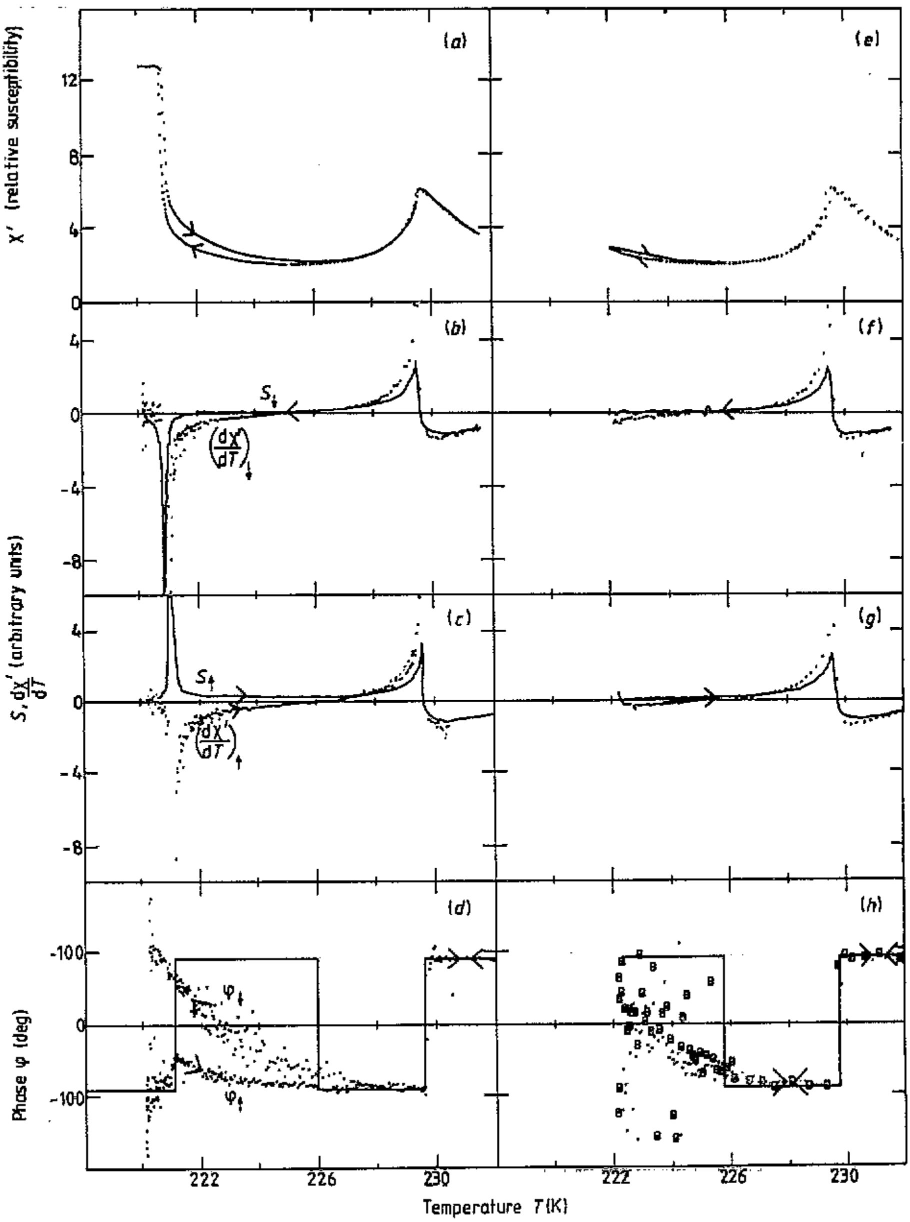


**Figure 3.** Comparison of the magnetic susceptibility $\chi(T)$ and temperature modulation $S \equiv \Delta\chi'(T)/\Delta T$ results for single-crystal terbium: (*a*)–(*d*) the results obtained on first cooling the sample to $T \simeq 220$ K (just below the ferromagnetic ordering temperature $T_C$) and then warming; (*e*)–(*h*) the equivalent results obtained on first cooling the sample to $T \simeq 222$ K (just above $T_c$). (*a*) and (*e*) show $\chi'$ as a function of temperature for an applied field of 3 A $m^{-1}$ RMS and $f = 1$ kHz. The $S$ data were obtained with $\Delta T \simeq 20$ mK and $f_m = 0.2$ Hz. Cooling and warming rates were about 0.1 K $min^{-1}$ or less. (*b*) and (*f*) show the analytical derivative $d\chi'/dT$ (●) and the temperature modulation signal $S$ (———) for the cooling (↓) experiment of (*a*) and (*e*) respectively. (*c*) and (*g*) are the equivalent warming (↑) curves. (*d*) and (*h*) show the cooling and warming phase $\varphi$ (of $S$) versus temperature. The phase expected of $d\chi'/dT$ (———) is shown for comparison. In (*g*) the points represent cooling data and the

been presented elsewhere (Wantenaar *et al* 1976, 1979, McKenna *et al* 1983). The variable-temperature cryostat and data-logging system used in these measurements have also been described (McKenna *et al* 1982) with information on the high-purity (99.98 wt% (99.7 at%)) terbium single crystal given by McKenna *et al* (1981a). The crystal was prepared using the metallothermic method (Beaudry and Gschneider 1978).

In order to obtain detailed information on the magnetic behaviour in the narrow AF region (about 8 K), an operating frequency $f = 1$ kHz was chosen for improved sensitivity. This operating frequency led to an electromagnetic skin depth attenuation in the susceptibility of, for example, about 10% for measurements at about 201 K in the ferromagnetic region, with smaller distortions in the AF temperature region. Furthermore, as indicated previously (McKenna *et al* 1981b), the temperature modulation signal also suffered attenuation due to a thermal skin effect even at a modulation frequency $f_m = 0.2$ Hz, the lowest available with our lock-in amplifier (LIA). A related consideration is that the raw $S(T)$ signals contain a contribution from $\Delta T(T)$, as well as $\Delta\chi'(T)/\Delta T$. Measurement of $\Delta T(T)$ can be used to determine the AC specific heat. Studies of the present sample indicated an anomaly of about 20% in $\Delta T(T)$ at $T_N$ and a smaller change at $T_C$ (McKenna *et al* 1981b). However, for the sample assembly used in the current susceptibility experiments for which primary and secondary coils are required, no anomalies in $\Delta T(T)$ (to within a variation of about 3% or more) were observed. Consequently no corrections for $\Delta T(T)$ variation have been applied to the $S(T)$ signals presented below. Because of these experimental limitations, only qualitative interpretation of the results is presented in section 5.

## 4. Results

Figure 3 shows a comparison between two sets of warming and cooling data for the magnetic susceptibility $\chi'$ and the simultaneously acquired temperature modulation signal $S$ for the single-crystal terbium sample around the AF region. In the first experiment (figures 3(*a*)–3(*d*)), the sample was cooled to a temperature $T \simeq 220$ K, just below $T_C$ ($\simeq$221 K), before being warmed back through the AF region. In the second experiment (figures 3(*e*)–3(*h*)) the sample was cooled to $T \simeq 222$ K (just above $T_C$) before again being warmed through the AF region.

For both experiments, $S$ and $d\chi'/dT$ are in general agreement for warming and cooling in the paramagnetic region (figures 3(*b*), 3(*c*), 3(*f*) and 3(*g*)). There is qualitative agreement between $S(T)$ and the temperature derivative of $\chi'(T)$ around $T_N$. In particular, $S$ reflects the expected 180° phase change in $d\chi'/dT$ associated with the peak in $\chi'(T)$ at $T_N$ (figures 3(*d*) and 3(*h*)). We attribute the lack of full agreement at this continuous transition to the incomplete correction for $\Delta T(T)$ as discussed above. For cooling through the AF region the temperature modulation signal is close to zero ($S_\downarrow \simeq 0$) with its phase attempting to track that expected from $(d\chi'/dT)_\downarrow$, the analytical derivative of

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boxes the warming data. Note that the maximum and minimum values for the off-scale peaks of (*b*) and (*c*), respectively, are

$$S_{\downarrow\min} = -13.8 \qquad (d\chi'/dT)_{\downarrow\min} = -139.5$$

$$S_{\uparrow\max} = +9.7 \qquad (d\chi'/dT)_{\downarrow\min} = -88.7.$$

Note also that the apparent differences between $\chi'$ for warming and cooling around $T_N$ are artefacts of rate dependence of temperature. The more careful measurements focusing on $T_N$ given in figure 1(*c*) of McKenna *et al* (1981a) show no hysteresis in $\chi'$ at $T_N$.

$\chi(T)_{\downarrow}$. There is qualitative agreement between $S_{\downarrow}$ and $(\mathrm{d}\chi'/\mathrm{d}T)_{\downarrow}$ at $T_C$ (figure 3(*b*)) only in that the sign of the two anomalies agree.

On warming from below the ferromagnetic ordering temperature $T_C$, $S_{\uparrow}$ is markedly different from $(\mathrm{d}\chi'/\mathrm{d}T)_{\uparrow}$; a peak of opposite polarity is observed at $T_C$ and an $S$ signal of polarity opposite to that of $(\mathrm{d}\chi'/\mathrm{d}T)_{\downarrow}$ continues well into the AF region (figure 3(*c*)). In contrast, for the experiment in which the sample was warmed from a start temperature above $T_C$ no such behaviour was observed and $S_{\uparrow}$ was indistinguishable from $S_{\downarrow}$ (figure 3(*g*)).

The behaviour of $S_{\uparrow}$, when the sample was warmed from a start temperature just below $T_C$, was essentially the same as that obtained on warming from a significantly lower start temperature (about 210 K) (see figure 4(*c*) of McKenna *et al* (1982)). Preliminary measurements on a polycrystalline sample of terbium (McKenna *et al* 1979) showed a similar but broadened behaviour. As already reported, temperature hysteresis of about

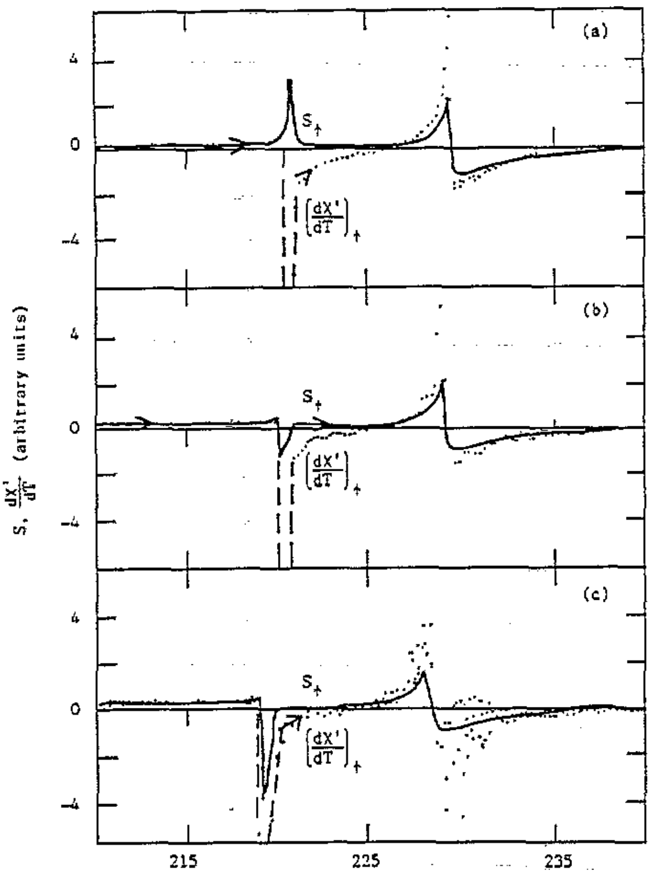


**Figure 4.** A comparison of the temperature modulation results $S_{\uparrow} = [\Delta\chi'(T)/\Delta T]_{\uparrow}$ (———) obtained on warming for various values of the temperature modulation amplitude $\Delta T$ and the corresponding analytical derivatives $\mathrm{d}\chi'(T)/\mathrm{d}T$ (•, – –): (*a*) $\Delta T = 65$ mK; (*b*) $\Delta T \simeq 130$ mK; (*c*) $\Delta T \simeq 250$ mK. Note that the minimum values for $(\mathrm{d}\chi'/\mathrm{d}T)_{\uparrow}$ for the off-scale peaks are $-17.3$, $-14.6$ and $-14.0$ for (*a*), (*b*) and (*c*), respectively.

0.25 K has been observed in the first-order transition at $T_C$ (McKenna *et al* 1981a). This hysteresis has been confirmed by the specific heat measurements of Jayasuriya *et al* (1983, 1984) on the same single-crystal sample.

Experiments were therefore carried out for a range of temperature modulation amplitudes up to peak values of $\Delta T \simeq 250$ mK in order to examine the effects of this thermal hysteresis. Apart from the inevitable shifting and broadening effects on the $S$ anomalies due to the increased temperature modulation amplitude, there was relatively little change in the behaviour of $S_\downarrow$ (McKenna 1980). However, figure 4 shows that, on warming, as $\Delta T$ is increased, there is a dramatic change in the response $S_\uparrow$ to the modulation cycle at $T_C$, resulting in the same polarity as $(d\chi'/dT)_\uparrow$ for $\Delta T \gtrsim 130$ mK. In addition, $S_\uparrow$ in the AF region reduces to approximately zero for increased modulation amplitudes, compared with the essentially invariant behaviour for $S_\uparrow$ around $T_N$ with increased modulation amplitude (figure 4).

## 5. Discussion

The essential agreement between $S_\downarrow$ and $S_\uparrow$ in the AF region for warming from start temperatures above $T_C$ (figures 3(*f*) and 3(*g*)) indicates that there is little change in the magnetic structure when the sample is cycled within the AF region. The smaller value of $S$ compared with $d\chi'/dT$ simply reflects the hysteresis in $\chi'$ (i.e. $\chi'_\uparrow > \chi'_\downarrow$) observed in figure 3(*e*). This hysteresis is also shown by the results obtained for $\chi'(T)$ in the absence of temperature modulation (figure 5(*a*)). This hysteretic behaviour is represented schematically in figure 5(*b*); for a cooling experiment the modulation cycle would follow cycle AB in figure 5(*b*) while for a warming experiment started from a temperature above $T_C$ it would follow the path CD. This behaviour in $\chi'$ and $S$ is similar to that for polycrystalline dysprosium (McKenna *et al* 1983). It is consistent with the model based on equation (1) whereby hysteresis in $\chi'(T)$, *when the sample temperature has not crossed* $T_C$, is due to hysteresis in the number of spins in thin AF domain walls.

Difficulty in separating effects linked with the microscopic magnetic structure changes at $T_C$ and the associated domain changes makes definitive interpretation of the behaviour of $S_\uparrow$ and $S_\downarrow$ at $T_C$ uncertain. The mere existence of anomalies in $S$ at $T_C$ on both warming and cooling, for modulation amplitudes *smaller* than the temperature hysteresis in $T_C$, is itself significant (e.g. figures 3(*b*) and 3(*c*)). Firstly, this indicates that the discontinuous first-order change in the microscopic magnetism, while dominant by volume (figure 3(*a*)), is not simultaneous over the entire crystal volume and, hence, is not in isolation of changes in technical magnetization processes. This follows from consideration of the response of an ideal first-order transition to such a low-amplitude thermal wave. Despite many cycles of the thermal modulation wave, only one transition through $T_C$ would take place owing to the hysteresis in $T_C$ and, as for AC specific heat measurements (McKenna *et al* 1981b), the integration provided by the LIA would yield no measurable anomaly. The existence of the $S_\uparrow$ signal at $T_C$ for small temperature modulation amplitudes (figure 3(*c*) and figure 4) and the fact that $S_\uparrow$ is of opposite sign to $(d\chi'/dT)_\uparrow$ at $T_C$ are clear evidence that the signal is not dominated directly by the microscopic magnetization changes. This result further demonstrates that, even though the anomaly observed in $S_\downarrow$ at $T_C$ on cooling (figure 3(*b*)) exhibits the behaviour and sign expected of $(d\chi'/dT)_\downarrow$, this $S_\downarrow$ anomaly must represent magnetization processes other than just the microscopic ones. Indeed, $S_\downarrow$ at $T_C$ is much smaller in magnitude than $(d\chi'/dT)_\downarrow$ (see caption for figure 3(*b*)) (McKenna 1980). Rather, because of the

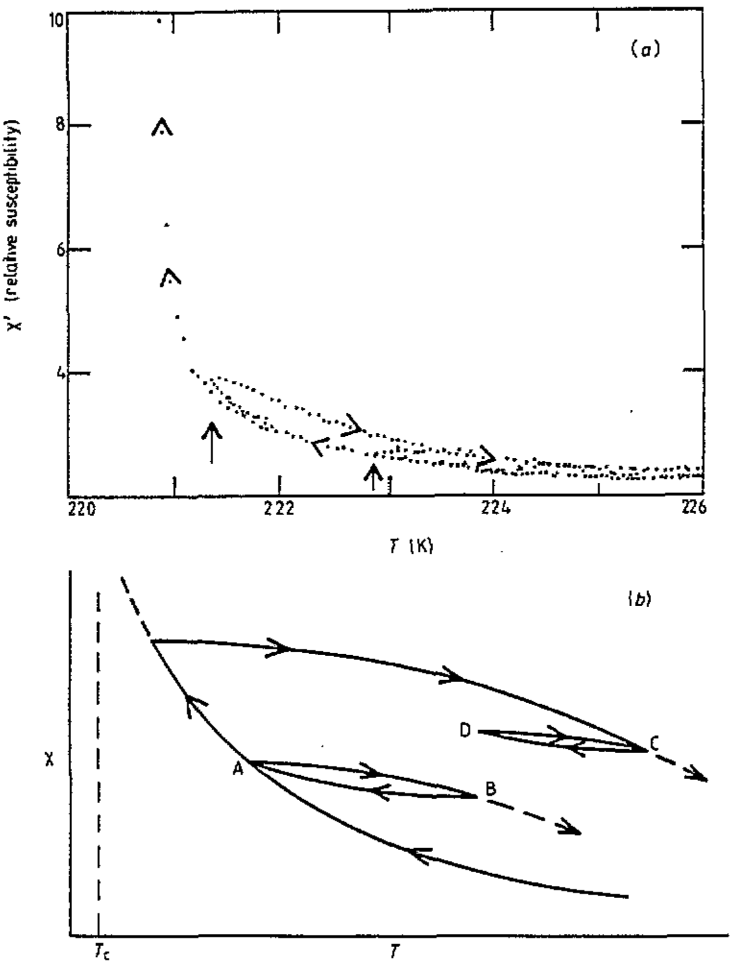


**Figure 5.** (*a*) The temperature dependence of the magnetic susceptibility $\chi'$ of terbium on cooling through the AF region ($H_{RMS} = 3$ A m$^{-1}$; $f = 1$ kHz). Also shown are the results of separate experiments in which the sample is cooled to and rewarmed from the temperatures 221.3 and 222.8 K indicated by the vertical arrows. (*b*) A schematic representation of (*a*), depicting cooling and warming sequences (without crossing $T_C$) and modulation cycles AB and CD.

ability of the hysteresis in $T_C$ to effectively dilute the idealized first order *microscopic* contribution to $S$ at $T_C$, these $S$ anomalies focus more on rearrangements in the complementary technical magnetization processes and in the magnetism of small regions that are *not undertaking the first-order transition*. The results for $S_{\uparrow}$, presented in figure 4, verify these biases for small modulation amplitudes. As expected, for modulation amplitudes for which $2\,\Delta T$ becomes greater than the hysteresis $h$ at $T_C$ ($h \approx 0.25$ K (McKenna *et al* 1981(b)), the $S_{\uparrow}$ anomaly reverts to the same polarity as $(\mathrm{d}\chi'/\mathrm{d}T)_{\uparrow}$. In this case the magnetic state of the sample is changed back and forward between the ferromagnetic and helical AF phases during each modulation cycle with $S_{\uparrow}$ now reflecting the resultant large changes in microscopic susceptibility over these $\Delta T$ amplitudes and the hysteretic contributions from the technical magnetization processes being relatively less important.

Another striking feature of the temperature modulation results, on warming through $T_C$ into the AF region, is the persistence throughout this region (221–229 K) of an opposite (positive) polarity $S_\uparrow$ signal, compared with $(\mathrm{d}\chi'/\mathrm{d}T)_\uparrow$ (figure 3(*c*)). This feature was also observed in the results for polycrystalline Dy (figure 1(*c*) (McKenna *et al* 1983)). For single-crystal Tb this effect persists above the temperature ($T \simeq 225$ K) at which ferromagnetic or intermediate domains and associated domain walls are no longer considered to contribute to $\chi'(T)$ (as indicated by the absence of transient enhancement

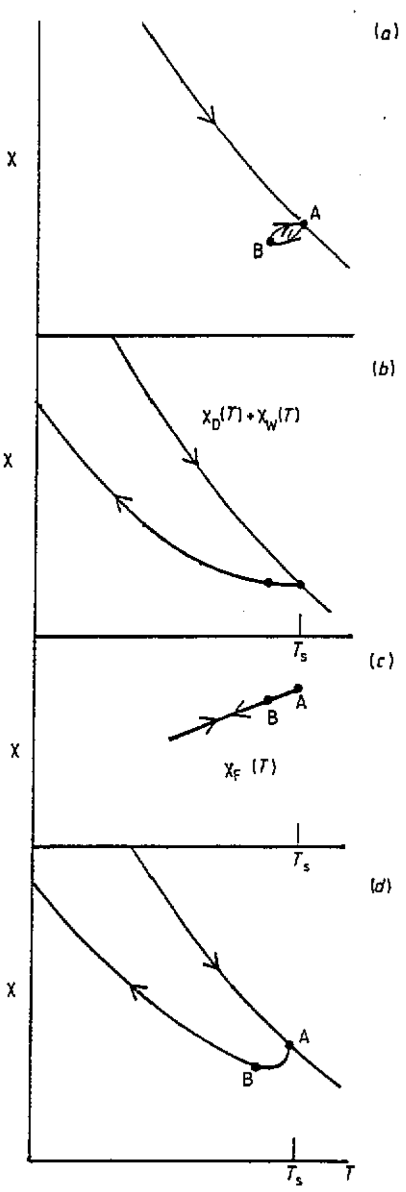


**Figure 6.** (*a*) A schematic representation of the behaviour of susceptibility in a temperature modulation cycle AB on warming from the ferromagnetic region to the AF region. Such an effect is expected when the observed polarity of $S_\uparrow \equiv (\Delta\chi'/\Delta T)_\uparrow$ is opposite to that of the analytical derivative $(\mathrm{d}\chi'/\mathrm{d}T)_\uparrow$. (*b*) An illustration of the hysteresis expected in $\chi_D(T) + \chi_W(T)$ for warming to a temperture $T_S$ in the AF region followed by recooling. Points A and B indicate the limits of a temperature modulation cycle. (*c*) A plot of the temperature dependence of the contribution to susceptibility of $\chi_F(T)$[or of $\chi_{WF}(T)$] as discussed in the text. (*d*) The predicted behaviour of $\chi'(T)$ using equation (8) where the contributing terms of the equation exhibit the temperature dependences shown in (*b*) and (*c*).

in $\chi'(T)$ above this temperature (McKenna *et al* 1981a)). This effect suggests behaviour in $\chi'$ which we label 'anti-hysteresis' with modulation loops of the form shown in figure 6(*a*). However, as the effect occurs only on warming from the ferromagnetic region it is clear that the AF region carries some memory of its previous ferromagnetic state.

Two types of such memory have been observed in neutron topographic studies of terbium. Firstly, Baruchel *et al* (1981) and Palmer *et al* (1986) observed differences in the structure of AF domains depending on whether cooling from the paramagnetic region or warming from the ferromagnetic region. The significant difference in the domain structure for the two methods of approach to the AF region was that a regular arrangement of domains was formed on warming from the ferromagnetic region, with a tendency for aligned stripe domains of long axis normal to the *c* axis, whereas randomly shaped domain structures were obtained on cooling from the paramagnetic region. Secondly, Baruchel *et al* (1986) observed that, on warming from below $T_C$, regions of enhanced scattering were observed which they explained as ferromagnetic islands embedded in the helimagnetic matrix, nucleated around magnetic impurities. A mass spectrometry analysis of our Tb single crystal (Ames Laboratory, private communication 1979) revealed 110 (at.) ppm Fe which would become spatially aligned in the ferromagnetic phase of Tb, tending along the easy *b* axis, the direction probed with the AC field. This extra contribution to the AC $\chi'$ from Tb islands of fan-like or ferromagnetic-like character should be relatively reproducible for a given sample. Note that, although the magnetic inclusions are required to be spatially aligned on warming through $T_C$, their ability to drive adjacent regions of Tb into a fan-like or ferromagnetic-like state should be of equal capability on cooling the crystal into the AF region. A second consideration is that some coexistence of ferromagnetic and AF regions in the sample near $T_C$, due to temperature gradients or residual impurities, is to be expected in real samples, even those of highest purity. It is difficult to distinguish between residual impurity and temperature gradient rounding of $T_C$ in a high-purity crystal such as that under study. Certainly, both figure 3(*a*) and our more detailed measurements of $\chi'$ for the same single crystal (McKenna *et al* 1981a) indicate persistent rounding in $\chi'$, immediately above the first-order transition region. Such behaviour is consistent with a spread in $T_C$ values and could reflect an enhanced contribution from quite small regions of the single crystal. We suggest that, on warming, recalcitrant regions of the sample which remain ferromagnetic above the average transition temperature $\bar{T}_C$ could serve as thicker AF domain walls of ferromagnetic character, locked between domains of opposite chirality. Unlike the contribution from magnetic inclusions, this effect may be less reproducible but would still be capable of giving an additional contribution to $\chi'_\uparrow$ compared with $\chi'_\downarrow$. Both the thicker ferromagnetic walls and the ferromagnetic islands, in essence, represent departures of the real sample from the ideal. We predict that, as sample quality improves further, $\chi'_\uparrow$ on warming through $T_C$ will approach $\chi'_\downarrow$.

The contributions to $\chi'(T)$ on warming from the ferromagnetic region can therefore be expressed by extending equation (1) such that

$$\chi'(T) = \chi_D(T) + \chi_W(T) + \chi_F(T). \qquad (8)$$

$\chi_W(T)$ is the AF domain wall contribution which is different from that for cooling, and $\chi_F(T)$ is the contribution due to ferromagnetic islands such as those observed by Baruchel *et al* (1986).

The greater value of $\chi'(T)$ on warming from start temperatures below $T_C$ compared with $\chi'(T)$ for cooling therefore requires either $\chi_W(T)_\uparrow > \chi_W(T)_\downarrow$ (del Moral and Lee 1974, McKenna *et al* 1980) or $\chi_F(T)_\uparrow > \chi_F(T)_\downarrow$, or a combination of both. The last two terms of equation (8) are magnetization processes that contribute heavily to providing the positive $S_\uparrow$ anomaly observed at $T_C$ for small modulation amplitudes, although on a volume basis they represent a far smaller fraction of the crystal compared with that volume which undergoes the discontinuous collapse from parallel moments to spiral spin in the basal plane.

One interpretation of the anti-hysteretic-type behaviour observed for $\chi'$ in a warming experiment which takes the sample from the ferromagnetic region to the AF region (e.g. figure 6(*a*)) is that the terms $\chi_D(T)$ and $\chi_W(T)$ in equation (8) behave in a similar manner as for cooling, so that the contribution to susceptibility from these two terms exhibits only simple hysteresis (figure 6(*b*)), while $\chi_F(T)$ is a slowly varying function of temperature with the opposite sense to that of the first two terms (figure 6(*c*)). When a sample is warmed from below $T_C$ to some temperature $T_S$ in the AF region (figure 6(*d*)) and then recooled from $T_S$, $\chi_D(T)$ and $\chi_W(T)$ are virtually constant so that $\chi'$ decreases owing to the effect of $\chi_F(T)$ (figure 6(*d*)). Since $\chi_F(T)$ represents the contribution to $\chi'$ from ferromagnetic islands, it would be expected to have the temperature dependence of $\chi'$ in the ferromagnetic region, such that

$$\chi_F(T) \propto \sigma(T)/K(T) \qquad (9)$$

where $\sigma(T)$ is the reduced magnetization. $K(T)$ would be expected to have a temperature dependence which is similar to that of the basal plane anisotropy term $K_6^6(T)$. $\chi_F(T)$ would therefore increase with increasing temperature because of the decreasing local anisotropy $K(T)$, thus providing the temperature dependence required to contribute to this apparent anti-hysteretic behaviour. This model explains the modulation cycle observed for the warming experiment shown in figure 3(*c*). The various contributions to this experimental result are indicated schematically by figure 6 in which the hysteretic behaviour expected of the separate components to $\chi'(T)$ in equation (8) are shown. The magnetic response as measured by $S_\uparrow \equiv [\Delta\chi'(T)/\Delta T]_\uparrow$ during a temperature cycle of modulation amplitude $\Delta T$ and oscillating between the points A and B, say, occurs as a result of the combined effects of susceptibility components which exhibit differing thermal hysteretic behaviour.

The suggestion that the modulation cycle may reflect a variation in the magnetic susceptibility $\chi'$ of the form shown in figure 6(*d*) is further supported by consideration of the larger-modulation-amplitude results for $S_\uparrow$ in the AF region. The reduction in the amplitude of $S_\uparrow$ in the AF region with increasing $\Delta T$ (figure 4) can be explained in terms of the modulation cycle changing from path AB of figure 7 for low modulation amplitudes, to path AE for higher modulation amplitudes, where $d\chi'/dT$ is approximately zero.

The second interpretation of the anti-hysteretic behaviour assumes that $\chi_F(T)$ is insignificant and that $\chi_W(T)_\uparrow$ can be expressed as

$$\chi_W(T)_\uparrow = \chi_{WS}(T) + \chi_{WF}(T). \qquad (10)$$

$\chi_{WS}(T)$ is due to walls of the type predicted from the energy calculations in section 2, and $\chi_{WF}(T)$ is an additional contribution due to thicker domain walls formed when the sample is warmed from the ferromagnetic region. If these walls were sufficiently thick for the majority of the spins in the walls to sense a ferromagnetic exchange coupling, then one might expect a local anisotropy field $K(T)$ to exist within these walls. Any response of these spins to an applied field would then be opposed by this local anisotropy, so that the susceptibility $\chi_{WF}(T)$ for such walls is somewhat similar to that of a single-domain ferromagnetic region. If we assume that $\chi_{WS}(T)$ exhibit hysteresis and that

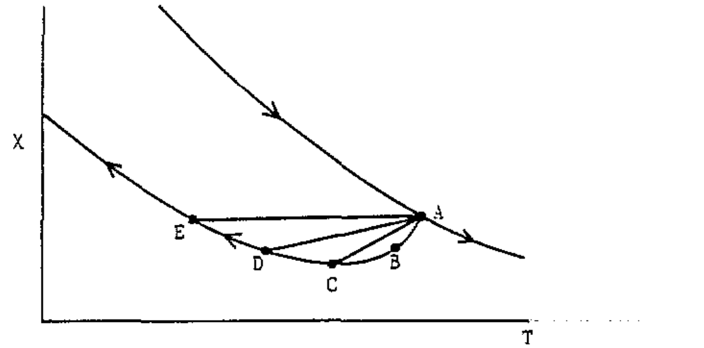


**Figure 7.** A schematic representation of the behaviour of $\chi'(T)$ on warming from below $T_C$, also indicating the change in behaviour obtained in temperature modulation experiments with modulation cycles of increasing amplitudes depicted, respectively, by AB, AC, AD and AE.

$\chi_{WF}(T)$ has the temperature dependence in equation (9) and figure 6($c$), then this, too, would cause $\chi'(T)$ to have the dependence in figure 6($d$).

In comparing these two explanations, the following points should be noted. Firstly, the absence of TE (the enhancement of $\chi'(T)$ by a transient bias field (Wantenaar *et al* 1976)) for Tb above $T \simeq 225$ K (McKenna *et al* 1981a) indicates the absence of ferromagnetic domain walls (i.e. walls separating regions of residual ferromagnetism) above this temperature. Therefore any ferromagnetic islands above $T \simeq 225$ K should be single-domain regions. This absence of enhancement of $\chi'$ is also consistent with the suggestion of thick AF domain walls of ferromagnetic character, in that such domain walls are equivalent to small single-domain ferromagnetic regions.

The second point to note is that comparison of figure 1 of Baruchel *et al* (1986) and figure 2($a$) of Palmer *et al* (1986) (apparently topographs of the same sample at the same temperature and at the same orientation) indicates that the observed ferromagnetic islands have the same orientation as the AF domain walls. It is also possible that these islands may be located at an AF domain wall, thus creating a locally thick domain wall, i.e. a combination of the two explanations given above. Finally, it should be noted that this discussion of the anomalous behaviour of the temperature modulation response $S$ in the AF region appears also to be applicable to our earlier results for dysprosium (figure 1($c$) of McKenna *et al* (1983)). No neutron topographs are, however, available for that material.

## 6. Conclusions

Our extension of the model of Thomas and Wolf (1965) for SS domain walls in the AF region of rare-earth metals predicts that the wall thickness should decrease with increasing temperature. This has led to improved understanding of the magnetic hysteretic and fine structural effects observed in the AF region of dysprosium and terbium following detailed measurements of their responses to an alternating magnetic field and a thermal wave.

On first cooling the samples from the high-temperature paramagnetic state to the AF region, the magnetic susceptibility and thermal modulation results obtained on subsequent warming of samples indicate a relatively small hysteresis in $\chi'(T)$ consistent with a decrease in the thickness of thin AF domain walls with increasing temperature. On warming the high-purity single-crystal; Tb from the ferromagnetic to the helimagnetic AF region, an extra positive contribution to the temperature modulation signal is observed throughout most of the AF region (figure 3(*c*)). This 'anti-hysteretic' behaviour in $\chi'(T)$ in the AF region is due to aligned ferromagnetic islands (centred around magnetic impurities) (Baruchel *et al* 1986) and/or to thicker domain walls of ferromagnetic character retained as a result of the imperfect first-order phase transition of the sample as a whole.

It is pointed out that, for small $\Delta T$ amplitudes, the temperature modulation signals $S$ at the hysteretic first-order phase transition $T_C$ provide a sensitive measure of changes in technical magnetization processes and remanent magnetization processes of those regions of the sample not undergoing the first-order phase transition. This feature complements $\chi'(T)$ measurements which are dominated by microscopic changes in the magnetic order–order transition occurring over the greater bulk of the sample. As a consequence, the temperature modulation technique provides considerable insight into the finer features of the transition and in particular the ultimate quality of the sample.

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