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Observation of chirality domains in terbium by polarized neutron diffraction topography

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Résumé. — Nous avons effectué la première observation de domaines de chiralité, dans un monocristal de terbium porté dans sa phase hélimagnétique, par topographies aux neutrons polarisés. Leurs formes sont fonction du cycle thermique subi par l’échantillon : des domaines en bandes à parois planes perpendiculaires à l’axe de l’hélice, distantes de 0,15 mm en moyenne, se forment lorsqu’il est réchauffé à partir de la phase ferromagnétique, tandis que des domaines à parois courbes se produisent quand il est refroidi à partir de la phase paramagnétique. Cette dernière configuration est reproductible pour un état donné de distorsion de l’échantillon.

Abstract. — We have carried out the first observation of spiral spin or chirality domains, in a single crystal of terbium in its helimagnetic phase, by polarized neutron diffraction topography. The different domain structures observed are a function of the thermal history of the sample. When the sample is warmed from the ferromagnetic phase one observes stripe domains with long axis perpendicular to the helical axis and width of ~ 0.15 mm. Cooling the sample from the paramagnetic phase produces more randomly shaped domain structures, which are reproducible for a given distortion state of the sample.

1. Introduction. — The heavy rare earth metals Tb, Dy and Ho, as well as some rare earth alloys such as Tb-50 % Ho and Gd-40 % Y have an antiferromagnetic helical phase intermediate between the high temperature paramagnetic and low temperature ferromagnetic phases. In the helimagnetic phase the magnetic moments lie normal to the six-fold axis, c, which is also the helix axis. In a particular basal plane sheet the moments are aligned ferromagnetically but the direction of magnetization rotates between successive Tb sheets by an angle θ. The turn angle, θ, decreases slowly with temperature from 20° at $T_N$ to 18° at $T_c$, where, for Tb, $T_N$ is approximately 226 K and $T_c$ ~ 219 K [1].

A spiral spin structure can be described by [2] :

$$S_j = \frac{1}{2} S e^{-i k \cdot R_j} m^+ + \frac{1}{2} S e^{i k \cdot R_j} m^-$$

$m$ being a unit vector in the direction of the magnetic moments in the plane of the origin. If the propagation vector $k$ is changed into $-k$, or if the sense of rotation along $k$ is reversed, the chirality of the spiral spin structure is reversed. A sample in the antiferromagnetic, helimagnetic phase can therefore be divided into spiral spin or chirality domains. Neighbouring domains will be associated with right handed and left handed rotation of magnetic moments. Theoretical discussions about walls separating these domains have been carried out by Thomas and Wolf [3] and Kléman [4]. Chirality domains were invoked to explain experimental anomalies in the temperature dependence of ultrasonic attenuation and elastic constants [5, 6], in magnetic susceptibility measurements on Dy [7] and Tb [8], and on torque measurements on Dy [9]. But none of these techniques could demonstrate conclusively the actual presence of chirality domains. The present paper describes their visualization and a preliminary investigation of their properties.

The neutron diffraction pattern of a helimagnet includes pairs of magnetic satellite reflections, $hk\ell_+$ and $hk\ell_-$ around the $hk\ell$ nuclear reflections [2]. These magnetic reflections occur at points in reciprocal space displaced from the nuclear reciprocal lattice point by $\pm k$, the propagation vector. Polarized neutron diffraction can distinguish bet-
ween domains having spirals of opposite sense. Several authors [2, 10-12] pointed out that when the scattering vector \( \mathbf{h} \) is parallel to the propagation vector \( \mathbf{k} \) of the spiral and to the incident beam polarization \( \mathbf{P} \), the contribution to one of the magnetic satellite peaks comes from one kind of chirality domains while the other kind of domain diffracts on the other satellite peak. This was used to measure the relative domain volumes in MnP [13] and ZnCr\(_2\)Se\(_4\) [14].

Polarized neutron diffraction topography [15] is not only able to give information about the overall volume of the two types of domain, but also to obtain images of the domains, allowing an investigation of their sizes and shapes. The basic experimental arrangement for neutron diffraction topography is schematized in figure 1: the single crystal sample is set for a chosen Bragg reflection, and a neutron sensitive photographic detector is placed in the diffracted beam, as near the specimen as possible in order to retain the best geometrical resolution whilst avoiding the direct transmitted beam. Local variations of the recorded scattered intensity are associated with defects or domains in the crystal.

**Fig. 1.** - Principle of topographic techniques: the sample \( S \) is set to Bragg diffract the \( hkl \) reflection, and a photographic plate \( P \) is put across the diffracted beam to record the scattered spot, i.e. an image of the crystal.

2. **Sample preparation and experimental techniques.**

Initially measurements were carried out on a Tb-50% Ho sample since this has a wide helimagnetic phase extending from 80 to 180 K. The results were however inconclusive due presumably to the poor crystal quality of the sample. Tb was chosen next, although it has a limited helimagnetic phase of \( \sim 7 \) K, since it was hoped that its good crystal quality would lead to large chirality domains. In addition the topographic technique requires samples of high quality for optimum performance.

The sample used to carry out this work was a high purity single crystal of Tb grown at the Centre for Materials Science, University of Birmingham, by electron beam melting followed by zone refining. The single crystal was then placed in a solid state electrotransfer furnace and held just below the melting point for 1000 hours [16]. The sample was spark cut from the rod and spark planed down to size using fine passes at the later stages to reduce surface damage. Finally the sample was etched in a 50/50 mixture of acetic and nitric acids.

The final thickness of the sample was 0.3 mm and its diameter 5 mm, the \( c \) axis being contained in the \( (0110) \) plane of the disc. A neutron diffraction investigation of the crystal quality indicated that the mosaic spread of \( (0001) \) lattice planes is very small \((<2')\) while that of \((hh20)\) planes is \( \sim 0.8'\). In this latter case, the rocking curve appears to be split into several peaks, associated with subgrains.

The present work was performed on the diffractometer S20 of Institut Laue-Langevin in Grenoble. This instrument, which is mainly dedicated to topography, is installed 6 m downstream from the end of a curved thermal neutron guide. The beam was monochromatized and polarized by a saturated Cu\(_2\)MnAl single crystal with a mosaic spread of 24', set for a 111 Bragg reflection. The wavelength used was \( \sim 1.5 \) Å.

As mentioned above, in the ideal arrangement to distinguish between chirality domains the diffraction vector \( \mathbf{h} \), the propagation vector \( \mathbf{k} \), and the incident beam polarization \( \mathbf{P} \) are all parallel. \( \mathbf{k} \) is parallel, in our case, to the six-fold \( c \) axis. \( \mathbf{h} \) is parallel to \( c \) when a 0001 reflection is used; on the other hand \( \mathbf{h} \) has to be horizontal on S20, as in most neutron diffactometers, so that \( \mathbf{P} \) must also be horizontal. The vertically polarized neutron beam, which passes undisturbed through the vertical magnetic field produced by \( V \), is then adiabatically rotated to be nearly parallel to \( c \) by the horizontal magnetic field of \( H \) (Fig. 2). This polarization can be inverted either by means of a Mezei-type single coil flipper (F in Fig. 2) or simply by a 180° rotation of the \( H \) magnet about a vertical axis. The polarizing and adiabatic rotation efficiencies were checked by adding a third magnet, identical to \( V \), after the \( H \) magnet on the neutron path, setting a second saturated Cu\(_2\)MnAl crystal in place of the

**Fig. 2.** - Obtention of horizontally polarized neutrons: a vertically polarized monochromatic beam is obtained by using the 111 reflection from a saturated single crystal of Cu\(_2\)MnAl(M). The polarization is adiabatically rotated to horizontal following the magnetic field (vertical in the V magnet and horizontal in the H one). The sense of the polarization is reversed either by means of a coil F or by rotating by 180° the H magnet around a vertical direction. The sample S is in the cryostat C.
specimen and measuring the flipping ratio $R$ thus obtained. Values of $R$ over 20 have been measured.

The sample was cooled by using a closed cycle helium refrigerator (« Displex ») equipped with a special tail to allow a sample to film distance smaller than 2 cm. We use, to photographically detect the neutrons, an X-ray dental film (« Kodak Periapical Ultra Rapide ») backed by a 10 $\mu$m thick $^{157}$Gd screen acting as (n-p) converter. Exposure times were about 12 hours. The resolution of a topograph in the present geometry is estimated to be about 0.1 mm.

3. Experimental results. — Figure 3 shows the topograph performed at $T \sim 240$ K, in the paramagnetic phase, when using the 0002 reflection ; contrast associated with the crystal defects can be observed. All subsequent topographs were carried out at a fixed temperature in the antiferromagnetic phase which was approximately 2 K above $T_c$. Topographs taken at this temperature of the 0002 peak and its satellite reflections, 0002$\pm$, are presented in figure 4. Before the topographs 4a to 4f were taken the sample had been cooled below $T_c$ and then allowed to warm to the chosen temperature. Figure 4a shows the central 0002 nuclear reflection topograph : no images associated with walls or domains are observable, and the defect images are very similar to those of the topograph shown in figure 3. Figure 4b, performed using the 0002$\_$. magnetic satellite reflection and a horizontal neutron polarization which we will call « horizontal minus » (the opposite sense being of course « horizontal plus »), shows alternate white and black bands running in a direction normal to the $c$ axis. The contrast of these bands is reversed on figure 4c, where the neutron polarization was « horizontal plus ». It then follows that these bands are domains of opposite chirality. Figure 4d shows the « 0002$\_$. horizontal minus » topograph : it is identical...
to figure 4c, that is the « 0002_., horizontal plus »
topograph. The simultaneous change in satellite and
polarization sense gives the same domain images,
as expected (Figs. 4d and 4e).

Let us note that the horizontal polarization P is not
exactly parallel to k and h, and the structure factor F
of the less diffracting domains is not actually zero
but nearly ; the simplified approach we are using is thus
a good approximation.

An 0002_ topograph on the same domain configura-
tion performed with vertical polarization (Fig. 4f)
shows a very faint contrast, which we presently do not
understand, between the two sets of domains.

When the sample is cooled from the paramagnetic
phase, the domain structure is very different, as can
be seen in figures 4g and 4h. Domain walls no longer
lie on (0001) planes, that is normal to the spiral axis,
but feature irregular shapes and no preferred orienta-
tion. Some correlation can be nevertheless found
between the defects observable on figure 4a and the
domains of figures 4g and 4h. Repeated sample cooling
from above T_N indicates that the domain structure of
figures 4g and 4h is nearly reproducible, whereas the
ones obtained when warming from the ferromagnetic
phase always present walls in the (0001) planes but
the location of these walls varies from one experiment
to the next.

The stabilization of these domains occurs at tempe-
ratures which are not in the immediate vicinity of the
Néel temperature : a topograph performed using the
0002_ satellite reflection (the presence of this Bragg
peak ensuring that the sample is still in the heli-
magnetic phase) and « horizontal minus » polarized
neutrons at T ~ (T_N - 2 K) shows only defect
images. The isothermal application of a magnetic
field of 0.12 T along c in the antiferromagnetic phase
does not modify the helimagnetic domain structure.
Similarly the domains are insensitive to the same field
while the temperature is cycled from the paramagnetic
to ferromagnetic phase. This is only to be expected
since the magnetic moments in Tb are rigidly confines
to the basal plane by a large two-fold anisotropy.
A magnetic field parallel to [1010] at least partially
destroy the helimagnetic order, provided the field is
sufficiently strong. To observe the effect the sample
was cooled into the antiferromagnetic phase in zero applied
field and the domain structure of figures 4g and 4h
obtained. A field of 0.07 T was then applied parallel
to a and removed. The initial configuration (Figs. 4g,
4h) is modified in the central region of the sample,
where stripe-type domains appear. The orientation of
the present sample prevented the application of a field
along [1230], the magnetic easy direction in the basal
plane.

The same sample was again investigated after it had
been slightly strained. The intensity diffracted on the
central peak was then enhanced, indicating an increas-
ed distortion of the sample. A topograph performed
when the sample was cooled from the paramagnetic
phase shows that the walls are still not confined in
(0001) planes, but the observed domain structure is
different from the previous one. On the other hand
when warming from the ferromagnetic phase the
domain structure is mainly composed of stripe
domains with walls lying, as before, normal to the
c axis.

4. Discussion. — Anomalous behaviour of the
elastic properties and the magnetic susceptibility
of spiral spin antiferromagnetic rare earth metals
have been attributed to the presence of helimagnetic
domains. The magnetic susceptibility of single crystal
Dy [7] and polycrystalline Tb and Dy [8, 17, 18] is
higher than expected theoretically when the samples
are cooled from the paramagnetic phase. This has
been attributed to a contribution to the susceptibil-
ity arising from a ferromagnetic moment of the heli-
magnetic domain walls. In addition the magnetic sus-
ceptibility in warming for the ferromagnetic phase is in
excess of that observed on cooling and this thermal
hysteresis has been thought to arise due to different
antiferromagnetic domain structures for the two cases,
giving rise to different volumes of domain walls.

In a similar manner, the elastic constant c_{33},
measured by propagating a compressional wave down
the hexagonal c axis, and the ultrasonic attenuation
a_{33}, depend, for Tb, Dy, Tb-50 % Ho and Gd-40 %Y,
on the thermal history of the sample [6]. As a sample
is cooled from above T_N both c_{33} and a_{33} behave
normally with temperature ; that is c_{33} increases
monotonically while a_{33} decreases slowly with decreas-
ting temperature. However as the sample is warmed
from below T_c the elastic constant c_{33} falls, immedi-
ately above T_c, while a_{33} rises dramatically at the same
temperature. Both parameters only recover their
normal behaviour close to T_N.

The present investigation provides clues for under-
standing both of these experimental investigations.
The actual existence of the helimagnetic domains in
rare earth metals is confirmed and it is evident that
the domain configuration is dependent on whether
the sample is cooled from the paramagnetic phase or
warmed from the ferromagnetic phase. It is not evident
that the domains are smaller in the latter case. There-
fore the increase in domain wall volume when the
sample is warmed from below T_c, as suggested by the
excess susceptibility, may well be due to domain walls
that are wider than those produced on cooling from
above T_N. Caution must nevertheless be exercised
at this stage as the domain structures may well vary
from sample to sample.

If we concentrate on the ultrasonic measurements,
the attenuation due to the magnetic domains can
originate from several sources. Firstly from a ∆E
effect related with a rearrangement of the magneti-
tization associated with the ferromagnetic domain
walls; secondly from a modification of the domain
wall thickness as the stress wave propagates through
the sample; and thirdly from a resonant interaction between the ultrasound and the domains when the domain wall separation is of the same order of the ultrasonic wavelength. The first mechanism is not thought to be important since the domain walls are only a few atoms thick [19] and, more important, the application of a stress along the hexagonal c axis will not modify domain walls with magnetization lying in the basal plane. The second mechanism will absorb energy from the ultrasonic beam since the domain wall will move to a higher energy state if the wall thickness is increased or decreased from the optimum value.

On the third case, our assumption is that the domain walls are partially pinned on crystal defects or impurities; they would vibrate in the potential well in sympathy with the driving ultrasonic field, producing ultrasonic attenuation [20]. This final case is particularly appropriate when the samples are heated from below $T_c$ since the domains are then of the order of 0.1 mm-0.4 mm thick, corresponding to ultrasonic wavelengths of 50-12 MHz. This is just the frequency range where high ultrasonic attenuation is observed when heating from the ferromagnetic phase. Domains of smaller widths may well be present, leading to attenuation at higher frequency, but the resolution of the present technique precludes their observation.

5. Conclusions. — We have used polarized neutron diffraction topography to perform the first visualization of chirality domains. Our preliminary experiments have shown that these domains have different shapes when coming from paramagnetic or ferromagnetic phases, but contrary to expectation their volumes are of the same order. The reproducibility of the domain structure when the sample was cooled through $T_c$ and its variation when the state of strain of the sample was changed suggest that this domain structure could be related to crystal defects. On the other hand the more regular arrangement we obtained when warming through $T_c$ is likely to be associated with the ferromagnetic domain structure. Work is proceeding to confirm these points, to investigate the behaviour of the domains under the application of a magnetic field and if possible to find a means to control these domains [14] which could lead to useful neutron polarizers.

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