X-ray topographic observations of 180° magnetic domain walls in (001) plates of nearly perfect iron–silicon alloy single crystals

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[Received 30 November 1990 and accepted 21 January 1991]

ABSTRACT

Contrary to generally-held views, 180° magnetic domain walls lying in the cube planes normal to (001)-orientation single-crystal plates of Fe + 3 wt% Si can be detected by diffraction contrast on X-ray projection topographs by use of conventional experimental arrangements. Conditions for good observations are explained with illustrations. The dominant contrast appears at intersections of the 180° Bloch walls with the X-ray exit surface of the specimen and is generated by small localized deformations at these outcrops. The contrast is at best very weak, being only about 10⁻² of that generated by 90° domain walls.

§ 1. INTRODUCTION

In the earliest application of X-ray topography to study internal magnetic domain structures (Polcarová and Lang 1962), and in all subsequent descriptions of X-ray topographic experiments on single-crystal plates of iron–silicon alloy known to the authors, the magnetic features observed have been 90° domain walls. Theoretical and experimental studies of these have been concerned with, for example, the zig-zag nature of 90° walls that transect (001)-orientation specimens (Polcarová and Kaczér 1967, Polcarová and Lang 1971), strain-fields at junctions of 90° walls (Miltat and Klémán 1973) and strain-fields associated with ‘fir-tree’ closure domains (Miltat 1976).

An extended 90° wall is equivalent to a coherent (101) reflection twin plane in a slightly tetragonal lattice of axial ratio 1 + 3λ₁₀₀, where λ₁₀₀ is about 2.7 × 10⁻⁵ in annealed Fe + 3 wt% Si. With Bragg planes having diffraction vector \( \mathbf{g} \) and for which \( (\mathbf{m}_3 - \mathbf{m}_1) \cdot \mathbf{g} \neq 0 \), where \( \mathbf{m}_3 \) and \( \mathbf{m}_1 \) are unit vectors along the magnetization directions in the domains separated by the 90° wall, the long-range effect of the wall is that of a tilt boundary. Like such, it generates strong X-ray diffraction contrast, which has been studied quantitatively (Polcarová and Brádler 1982). On the other hand, 180° walls, which lie in (100) and (010) within an (001)-orientation plate, produce no long-range deformation, and on that account are expected to produce no X-ray diffraction contrast. However, these 180° Bloch walls do coincide with a thin sheet (effective thickness \( \approx 100 \) nm) that is under elastic stress as a consequence of the rotation of the magnetization direction (and hence of magnetostriction) occurring in the wall. At surface outcrops this stress relaxes. Small and highly localized though the resulting deformations are, the possibility of their producing detectable X-ray diffraction contrast should not be dismissed. The authors kept a watch for such contrast, and were
rewarded. The results described below date from the Summer of 1968. Display of some observations, with a few notes on technique, is deemed opportune now on two accounts: firstly to dispel the widely held belief of X-ray topographic undetectability of 180° walls (e.g. Kléman 1983), and secondly, to stimulate new experiments, taking advantage of the intense synchrotron X-ray sources and more refined topographic techniques now available.

§2. EXPERIMENTAL

Single-crystal rods, diameter 13 mm, were grown by zone-melting followed by long annealing (Kadečková and Šesták 1969). Plates parallel to (001) were prepared by spark machining and final chemical polishing. They contained sub-grains, diameters 2–5 mm typically, with misorientations at sub-grain boundaries generally in the range 10–60 arc seconds. Specimens were mounted so as to be free from elastic deformation. Many sub-grains had densities of random dislocations less than 10 lines mm⁻²; these provided the best environments in which to observe 180° walls. Other conditions satisfied in order to provide stress-free conditions for development of long segments of
Observations of magnetic domain walls

(a) X-ray projection topograph of (001) plate, printed with normal contrast, showing features producing strong diffraction contrast: low-angle boundaries, dislocations, 90° domain walls transecting the specimen and fir-tree-type surface closure domains. Direction [010] points upward, [100] to the right. Specimen thickness, t, is 110 \( \mu \)m, field height is 1.35 mm. Radiation MoK\( \alpha \), reflection 2\( \theta \) symmetrically transmitted, Bragg angle \( \theta_b = 14.52^\circ \). Horizontal dimensions compressed by \( \cos \theta_b = 0.97 \). Normal absorption exponent \( \mu t \sec \theta_b = 3.4 \). (b) Key to bulk domain structure imaged in projection topograph (a). Outcrops of domain wall boundaries on the X-ray exit surface of the specimen are indicated thus: 90° walls, continuous lines; 180° walls, interrupted lines. Surface closure domains omitted.

180° walls accurately parallel to cube planes were freedom from inclusions and all surface damage. 180° walls were detected in many specimens, and with specimen thicknesses ranging from 30 \( \mu \)m to over 100 \( \mu \)m. Diffraction geometries successful for observation of 180° walls parallel to (100) in a (001) plate included symmetrical transmission using reflections from (100), (110) and (110), and asymmetric transmission using reflections from (101) and (101) inclined at 45° to the specimen surface. Using MoK\( \alpha \), radiation, with which the best images were obtained, the 50 \( \mu \)m thick Ilford L4 nuclear emulsion plates were exposed sufficiently long to give (after low-temperature development) a high optical density of not less than about 1.5 in perfect-crystal regions, in order to enhance detectability of small changes in diffracted intensity. Whenever
good conditions for wall visibility were obtained, such as long wall segments running through dislocation-sparse regions, the topograph was immediately repeated. As long as no domain movement had occurred, this provided two identical high-density images. These were photomicrographed identically on to sheet film, and the two films were superimposed precisely for making high-contrast prints such as shown in figs. 2 and 3.

The topographs in figs. 1–3 are similarly oriented: [010] points upward, and [100] to the right. The view is towards the X-ray source, looking along the diffracted beam direction. Horizontal compression of the image becomes noticeable when the angle between diffracted beam and specimen normal is not small, and is evident in fig. 2. Figure 1(a) reproduces the topograph image with fairly faithful contrast. Features

Fig. 2

High-contrast micrograph of X-ray topograph image of part of field shown in fig. 1. Height of field 0.9 mm. MoKα, reflection 101 from Bragg planes inclined 45° to specimen surface. Projection of diffraction vector points to right. Bragg angle $\theta_B = 10.21^\circ$, diffracted beam makes $45^\circ - \theta_B = 34.79^\circ$ to specimen surface normal, so horizontal compression factor in image is $\cos 34.79^\circ = 0.87$. Images of two vertical 180° walls are seen: on left above the arrow, and on right as the vertical diagonal bisector of square (rhombus in projection) formed by X-ray exit surface outcrops of 90° walls.
Images of X-ray-exit-surface outcrops of 180° walls lying in (100) observed in asymmetric transmission topographs. MoK\textsubscript{2} radiation. Reflection 202. Projection of diffraction vector points to left. Bragg angle $\theta_b$ is 20°76', diffracted beam makes 45° - $\theta_b = 24°24'$ with specimen surface normal. Horizontal dimensions compressed by $\cos 24°24' = 0.91$. Specimen thickness similar to that in fig. 1. Magnification same as in fig. 2. (a) Walls visible between decoration by fir trees, in centre and in top right of field. (b) A wall free from fir trees, lying along vertical bisector of field.
running through the plate give sharp images when close to the X-ray exit surface and
diffuse images when close to the X-ray entrance surface. This effect is a notable feature
of the dislocation images. The superficial fir-tree structures present, e.g. in top right
corner and on the lower right, give sharp images and hence lie on the X-ray exit surface.
A sub-grain boundary intrudes along the right-hand edge, and another crosses the left-
hand lower corner, between the long 90° walls identified in fig. 1(b). The 90° walls
transecting the specimen lie parallel to (110) and (110) in the mean, but they have zig-
zag structure (which can be seen in more oblique views (Polcarová and Lang 1971)).
The black–white contrast in the butterfly lobes attached to junctions of 90° walls
(Miltat and Kléman 1973), and in the diffuse images of dislocations, is typical under the
absorption conditions applying (μt equal to a few units). In this topograph, as in other
reflections from planes containing [010], only the 180° walls parallel to (100) are
detectable. In fig. 1(a) such walls, which run vertically, can be seen on the original
topograph plate, and can be recognized on the micrograph print, especially by sighting
along the wall images at grazing incidence. These same 180° walls are more evident, but
still appear very weak, in the high-contrast print of the obliquely transmitted 101
topograph image reproduced in fig. 2. The diffraction geometry of this oblique
transmission determines that images of the X-ray exit surface outcrops of 90° and 180°
walls appear to the right of their entrance surface outcrop images in fig. 2. It is seen thus
that the 180° walls are detectable only along their X-ray exit surface outcrops in this
image.

Another pair of very-high-contrast micrographs is displayed in fig. 3. They show
different 180° walls, located about 3 mm distant from the field of fig. 1, and in a region of
similar thickness. Again only the exit surface outcrops of the 180° walls are evident, but
the width and contrast of their images in these second order, 202, reflections are
patently greater than in the first order, 101, shown in fig. 2. The 180° wall outcrop
images in both figs. 2 and 3 consist of a narrow line of excess intensity alongside a
broader and stronger line of reduced intensity. Estimates of the width of the deficiency
lines are about 3 μm in fig. 2 and about 7 μm in fig. 3. In the symmetrically transmitted
200 reflection, excess and deficiency in the 180° wall outcrop image more nearly
balance, and the width of both lines is about 2 μm, near the topographic resolution
limit. In all situations studied, the direction that crosses the wall outcrop image from
deficiency to excess points parallel to g, or to its projection on the specimen plate.

§ 3. Conclusions

There is a report of visibility of a 180° wall on X-ray topographs of an iron whisker,
axis [001] (Nagakura and Chikaura 1971). The case concerns a 180° wall tied to a Y
junction with a closure domain at the whisker tip. Visibility only occurred under
application of a magnetic field parallel to the whisker axis. This distorted the closure
domain and caused the 180° wall, tied at one end, to incline away from its stable (010)
orientation. The significant strength of the wall image, a broad deficiency line, reflects
the very different magnetic situation from that described above. It should be
emphasized that the observations reported here apply to ideal magnetic domain
structures in large, nearly perfect crystals, under conditions of freedom from
mechanical stress and from external magnetic field other than ambient.

From visual comparison of optical densities, it is estimated that the strongest 180°
wall outcrop images observed show contrast not more than about 1% of that of 90°
walls under similar diffraction conditions, taking the maximum departure of diffracted
intensity from that given by adjacent perfect crystal matrix as the contrast measure.
The elastic and magnetic conditions at outcrops of 180° walls in thick (001) plates have been discussed by Kroupa and Vagera (1969) and by Hubert (1971). The former work predicts lattice tilts exceeding some tenths of a second of arc, and of opposite sense, in volumes a few micrometres in width and depth on either side of the wall outcrop. Such could just be sufficient to give the diffraction contrast observed, and the excess/deficiency sequence is in the direction expected from diffraction theory (Penning and Polder 1961). X-ray topographic experiments hold some promise for checking models of 180° wall outcrops. Clearly, however, the experiments reported here have pushed conventional X-ray topographic procedures to the limit. Progress must lie with application of higher X-ray fluxes to build up improved signal-to-noise ratios, and of highly collimated monochromatic probing beams.

REFERENCES

MILTAT, J. E. A., 1976, Phil. Mag., 33, 225.