Advanced Materials '93, I / B: Magnetic, Fullerene, Dielectric, Ferroelectric,
Diamond and Related Materials, edited by M. Homma et al.
Trans. Mat. Res. Soc. Jpn., Volume 14B

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USING EXTREME FRINGE CONTRAST BEHAVIOR TO DETERMINE 180° DOMAIN POLARIZATION CONFIGURATION IN LEAD TITANATE SINGLE CRYSTALS

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180° domain boundaries in lead titanate crystals show intriguing extreme (outermost) fringe contrast behavior. Beside the typical α-type fringe behavior, some bright-dark alternatively arranged fringes were frequently observed. In the present work, an analysis based upon the two beam dynamical theory was conducted to predict the geometric and polarization configuration of a 180° domain boundary using the extreme fringe contrast (EFC) behavior. Employing different reflection vectors, the EFC of 180° domain boundaries varies. This has been exploited to determine the polarization vector arrangement at two sides of the boundary, and therefore the whole polarization configuration can be uniquely determined.

1. INTRODUCTION

There are relatively few reports on the $PbTiO_3$ domain boundary structures mainly due to the difficulty of growing crystals and its similarity to $BaTiO_3$. It is reported that $PbTiO_3$ crystals, with larger polarization and tetragonality, show very stable domain boundaries under an electron beam[1], and may present more significantly minute features due to its tetragonality [2]. TEM investigations on $BaTiO_3$ made by Tanaka and Honjo [3] have shown the basic configuration of 90° and 180° domain boundaries. Gevers et al. [4,5] applied the dynamical diffraction theory to calculate the fringe behavior of both inclined α-type and δ-type boundaries. Malis and Gleiter [6], using boundary contrast analysis, reported that both 90° and 180° domain boundaries consist of stacking-fault-type displacements in polycrystalline $BaTiO_3$. Accordingly, the displacement for a 90° boundary consists of a dilatation and shear of {110} planes and for a 180° boundary it lies approximately parallel to [101] and with greater magnitude. 180° domain boundaries in lead titanate crystals show intriguing extreme (outermost) fringe contrast behavior. Beside the typical α-type fringe behavior, some bright-dark alternatively arranged fringes were frequently observed. In the present work, we report the fringe contrast behavior of inclined 180° domain boundaries, and a method to determine 180° domain boundary geometric arrangements and polarization configuration using extreme fringe contrast (EFC) analysis.

2. EXPERIMENTAL PROCEDURE

The crystal growing procedure and specimen preparation method please refer to our previous work [7,8]. TEM investigation was conducted using a Hitachi H800 and a Jeol 2000FXII microscope with a double-tilt specimen holder operated at 200KV. The investigation employed different reflections at two-beam conditions and used mainly fringe contrast analysis. Different g vectors to reverse the EFC of the same boundary were employed to identify the displacement vector at the boundary and therefore construct the unique polarization arrangement for the domains.

3. RESULTS AND DISCUSSION

Figure 1 shows a scanning electron micrograph of a shell-like layered 180° domain arrangements in a good quality PbTiO3 crystal. Figure 2 shows bright- and dark-field TEM micrographs of inclined 180° domain boundaries which present fringe contrast at domain boundary positions. The pictures present the EFC behavior of a layered 180° arrangement corresponding to figure 1, which shows alternatively reversed contrast behavior at each boundary, indicating varied displacement vectors present at the boundaries. An analysis based upon the two beam dynamical theory was conducted and a rule similar to stacking-fault contrast analysis has been established to predict the geometric configuration of a 180° domain boundary using the extreme fringe contrast (EFC) behavior. The results are shown in Table I [7,8].

To determine polarization vector uniquely, we must know the displacement vector, and then the relation between the displacement and the polarization vectors. It is no doubt that the displacement are produced by the interaction of the polarization vectors. In the present case, we use the 180° domain boundaries shown in figure 2(b). In this picture, we see the arrangements of 180° domain walls, the contrast behavior of boundaries, and the diffraction vector **g**, which is 011. If we use different diffraction vector, **g**=$01\bar{1}$, to image, the results are shown in figure 3. The arrangements of domain walls are the same but the extreme fringe contrast was reversed. Note that the diffraction vector is 90° from the one in figure 2. Using Table I, one can determine the geometric arrangements of domain walls and one finds that the results using figures 2 and 3 are the same. Using tilting experiment, the arrangements of the walls in figures 2 and 3 are confirmed, as shown in figure 4. Let's take a region to construct the polarization arrangements of domains, as circled in figure 2. Since Malis and Gleiter [6] have reported that the displacement on 180° domain walls in $BaTiO_3$ is of <110> type. Our results (not shown) indicate that the same situation is followed in $PbTiO_3$ except that the magnitude of the displacement is larger. If we analyze the arrangement of domain walls, diffraction vectors and the displacement vector in a three dimensional way, shown in figure 5, one find that the displacement vector lies on a plane normal to a 020 plane and the fringes become extinct when using **g**=020. The displacement vector determines the contrast of a domain boundary using a specific diffraction vector. Since the displacement is produced by the interaction of two adjacent polarization vectors, the arrangement of polarization vectors will influence the resultant displacement. From the extinction conditions [7,8] and the domain boundary model proposed by Malis and Gleiter, one can derive a relation between the polarization and the displacement vectors. Figure 6 shows possible displacement vectors for two 180° polarization arrangements. Since **R** is normal to **g**=020, **R** must lie on a plane parallel to 020, and polarization vectors are along c direction, as shown in figure 6.

Ordinarily, we determine the c axis using diffraction pattern, the only ambiguity is to determine the direction of the displacement vector (either (a)/(b) or (c)/(d) in figure 6). This problem can be resolved as follows. Considering the extreme fringe contrast shown in figures 2 and 3, the contrast behavior is reversed by changing diffraction vector. This must be related to the sign of α (the phase shift) value. If we concentrate on the bracketed region shown in figure 2 and compare Table I, figures 2, 3 and 6, the displacement vector can be identified as shown in figure 7. Since the boundary is extincted using a 020 vector, and **g.R** is positive in figure 2 and is negative in figure 3, the displacement vector is therefore unambiguously determined as R=[$\bar{1}$01] in this case. So the polarization configuration around this boundary can be determined according to figure 6, as the arrangement in case (a). Since we know the boundary is inclined from upper right to lower left, the polarization vector is uniquely determined, shown in figure 7(c). Based upon this, the polarization arrangements in figure 2 and 3 can be determined unambiguously, as shown in figure 8. It appears that the present model not only identify the geometric arrangements but also the polarization configuration using extreme fringe contrast behavior.

4. CONCLUSIONS

An analysis based upon the two beam dynamical theory was conducted to predict the geometric and polarization configuration of a 180° domain boundary using the extreme fringe contrast (EFC) behavior. Employing different reflection vectors, the EFC of 180° domain boundaries varies. This has been exploited to determine the polarization vector arrangement at two sides of the boundary, and therefore the whole polarization configuration can be uniquely determined.

ACKNOWLEDGEMENTS

The authors are grateful to Professor D. A. Payne for providing part of crystals and valuable discussions. This project is supported by the Air Force Office of Scientific Research under Contract AFOSR-90-0174, U.S.A. and by the National Scientific Council under Contract NSC 82-0404-E-011-118T, Republic of China.

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Table I Table showing nature of the first and last fringes of an inclined 180° domain boundary for all possible arrangements. (B: bright; D: dark; diffraction vector to the right.)

	Arrangement	g*R	BFI	CDFI
1	g	>0	B B	D B
2	g	<0	D D	D B
3	g	<0	D D	B D
4	g	>0	B B	B D

B: bright; D: dark; diffraction vector to the right.

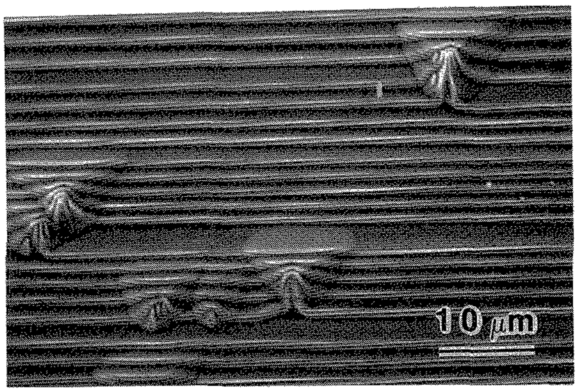

Figure 1 A scanning electron micrograph of a shell-like layered 180° domain arrangements in a good quality PbTiO3 crystal.

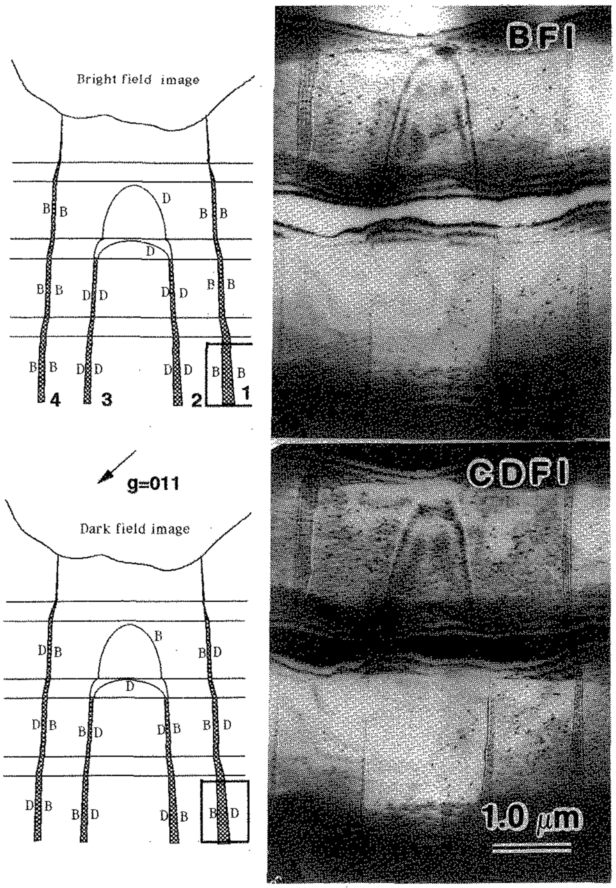

Figure 2 TEM micrograph showing a 90° and 180° domain boundary intersecting configuration, corresponding to figure 1, showing complicated fringe contrast behavior.

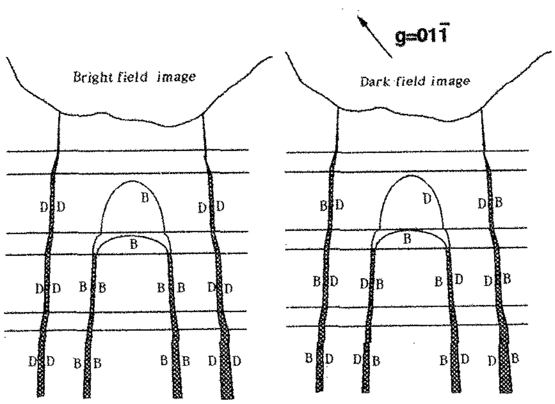

Figure 3 A schematic representation of the contrast distribution of the same domain boundary arrangement as in figure 2, but with different diffraction vector and showing reverse contrast behavior as figure 2.

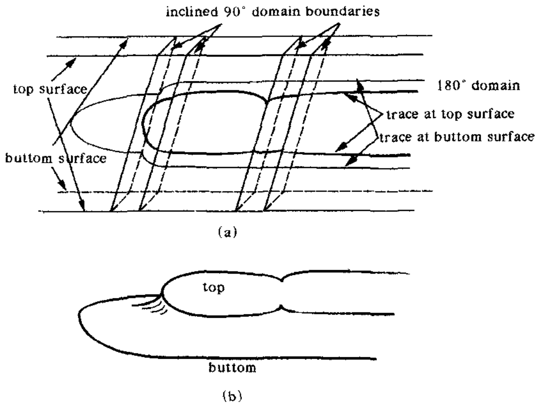

Figure 4 Schematic representation of the domain boundary configuration shown in Figures 2 and 3. (a) A three-dimensional scheme; (b) projction view of (a).

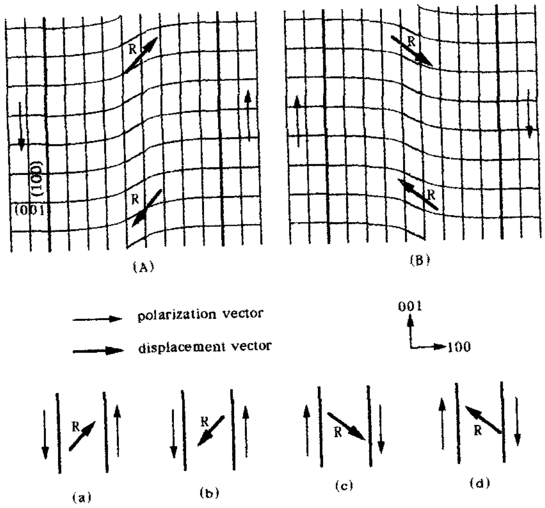

Figure 6 Possible displacement vectors for two 180° polarization arrangements.

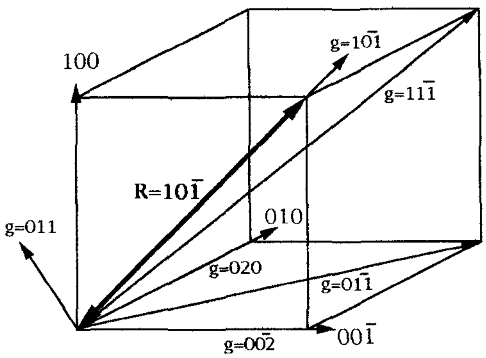

Figure 5 180° domain displacement vector analysis using extinction conditions with different reflections. The displacement vector is of <110> type.

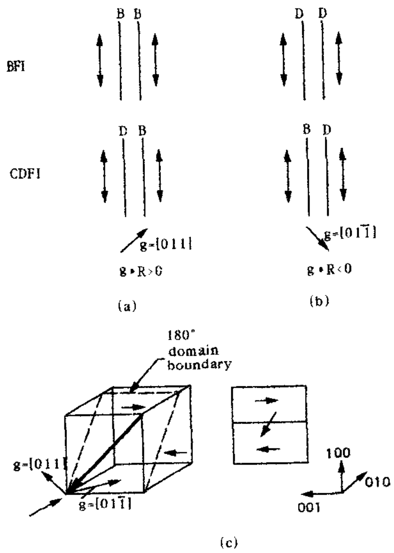

Figure 7 Displacement vector determination using extreme fringe contrast behavior in a three dimensional construction. The region bracketed in figures 2 and 3 is using as an example. See text.

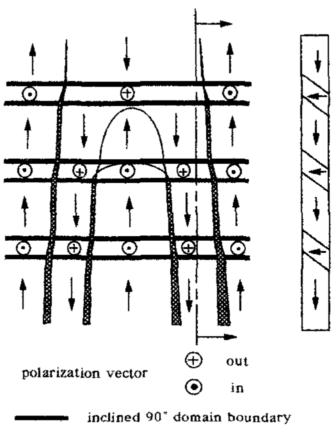

Figure 8 Polarization configuration of figures 2 and 3 derived from polarization vector determination.