ANALYSIS OF CHEVRON TWIN TILING OF $LaAlO_3$

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If cooled through the improper ferroelastic $Pm3m$ - $R\bar{3}c$ phase transition at 544 °C, single crystals of $LaAlO_3$ develop characteristic twin domains with {100} and {110}domain walls referred to the pseudocubic lattice. The resulting domain patterns on a $(100)_{\text{pseudocubic}}$ cut and polished surface of a $LaAlO_3$ wafer plate can be investigated in a quantitative way with scanning force microscopy (SFM). The ferroelastic deformation corresponds to a compression along one of the four <111> directions and extension perpendicular to it. From the rhombohedral angle of the pseudocubic unit cell $\alpha = 90.096(1)°$, the strain coefficient at room temperature is found to be $e = 8.4(1) \,.\, 10^{-4}$. For the calculation of matrices of the orientation variants each strain tensor has to be rotated towards the {100} planes of the cubic paraelastic phase. The resulting 24 matrices describe the real orientations of the domain states in ferroelastic twinned $LaAlO_3$ crystals.

1. Introduction

The ferroelastic phase transition of $LaAlO_3$ and its twining at room temperature have been subject of many investigations [1-5], especially since single crystals are used as HTSC- substrates. $LaAlO_3$ has a perovskite structure [6], the space groups of the high temperature paraelastic and the room temperature ferroelastic phase are $Pm3m$ and $R\bar{3}c$, respectively. According to the notation of Aizu [7], the phase transition of $LaAlO_3$ at 544 °C is labelled as the species $m3m\,F\bar{3}m$. Because of thermal stresses and imperfections in $LaAlO_3$ single crystals, different twin domains occur below the transition temperature. The number of domain states can be easily determined by the index of orders between the high and the low temperature symmetries [7]. As already pointed out in [8], the domain states have to be rotated with respect to the orientation of the paraelastic crystal lattice to be described in their real orientational relationship. In this paper we give a description how to derive the rotated domain states for the species $m3m\,F\bar{3}m$, i.e. the orientation variants for $LaAlO_3$.

2. Domain states, domain walls and rotation variants in $LaAlO_3$

According to Aizu [7] the maximal number of ferroelastic domains states in a twinned crystal can be calculated by the index of the point group symmetry orders between the paraelastic and the ferroelastic phase. In the case of $LaAlO_3$ the point group of the paraelastic high temperature phase is $m3m$ with the order 48 and the point group of the low temperature phase is $\bar{3}m$ with the order 12, hence the index 4 is the number of possible domain states. A concept to calculate the strain tensors of the domain states for every possible ferroelastic phase transition is also given by Aizu [9]. The strain tensors of the four domain states of $LaAlO_3$ are given in Table 1.

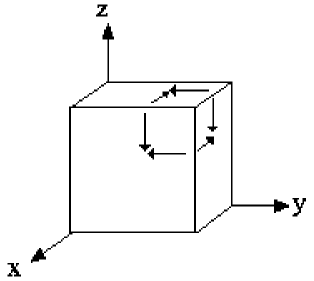

Fig. 1. Illustration of the lattice shear distortion for domain state I.

Fig. 1 shows a scheme of the distortion for domain state I, where every arrow marks a component of the strain tensor. Each component of the strain martix represents the distortion with respect to x, y and z. Because of the symmetric distortion of $m3mF\bar{3}m$ phase transitions, all strain components have the same absolute value which is $e = 8.4 \,.\, 10^{-4}$ for $LaAlO_3$. For cubic → rhombohedrral phase transitions

$$e = \frac{\sqrt{3} - \sqrt{3 + 6\cos\alpha_r}}{2\sqrt{3}} \tag{1}$$

where α_r is the rhombohedral angle of the pseudocubic lattice; in $LaAlO_3$ $\alpha_r = 90.096(1)°$. The condition, that the lattice strain in a twin wall must be equal on both sides in the two adjacent domains determines the crystallographic orientation of the twin wall [10]. To calculate the coordinates x_i of the possible twin planes between the domain states, the sum of all strain tensor elements are equated

$$\sum_{i,j}\left(e_{ij\,A} - e_{ij\,B}\right).x_i x_j = 0 \tag{2}$$

where the e_{ij} (A, B) are the strain tensor elements of the adjacent domains A and B, respectively. For any two domain states equation (1) delivers two mutually exclusive solutions. For the species $m3m\,F\bar{3}m$, twin walls are $\{100\}_{pc}$ and $\{110\}_{pc}$ as shown in Table 2. The subscript *pc* indicates that the Miller indices refer to the pseudocubic lattice of $LaAlO_3$.

Fig. 2 shows the SFM image of a twinned $(001)_{pc}$ surface of $LaAlO_3$ with all 4 domain states and the permissible domain walls between them.

Another approach to find the orientation of the twin planes is to look at the mirror planes which have been lost during the phase transition. These mirror planes likewise

Table 1: Twin-domain states of $LaAlO_3$ with the corresponding unique triad (compressional axis) and strain tensor [5].

Domain state	I	II	III	IV
Compressional Axis	[111]	$[1\bar{1}1]$	$[\bar{1}11]$	$[11\bar{1}]$
Strain Tensor	$\begin{pmatrix} 0 & \bar{e} & \bar{e} \\ \bar{e} & 0 & \bar{e} \\ \bar{e} & \bar{e} & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & e & \bar{e} \\ e & 0 & e \\ \bar{e} & e & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & e & e \\ e & 0 & \bar{e} \\ e & \bar{e} & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & \bar{e} & e \\ \bar{e} & 0 & e \\ e & e & 0 \end{pmatrix}$

Table 2: Allowed mutually exclusive twin domain walls between each pair of twin-domain states I to IV as given in Table 1.

I, II	(101), (010)
I, III	(011), (100)
I, IV	(110), (001)
II, III	$(1\bar{1}0)$, (001)
II, IV	$(01\bar{1})$, (100)
III, IV	$(10\bar{1})$, (010)

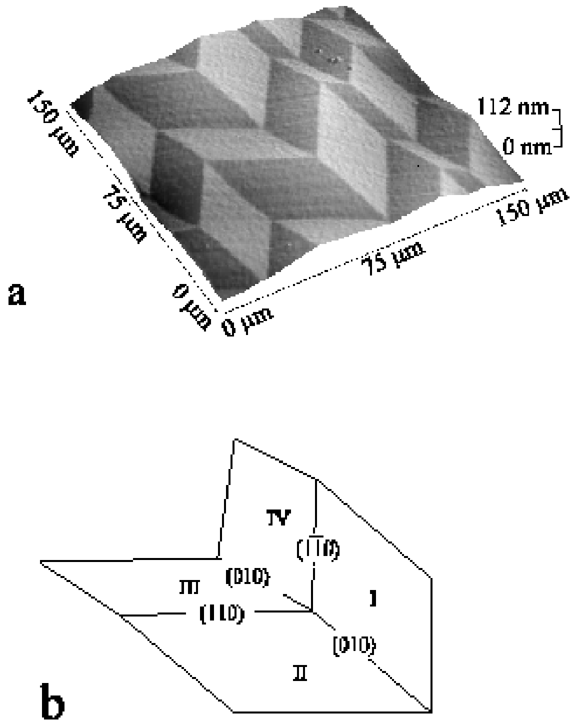

Fig. 2. (a) SFM image of the $(001)_{pc}$ surface of $LaAlO_3$ showing all permissible domain walls, (b) Schematic indication of domain states and domain walls, the kink in the (010) domain wall is due to the interaction of (010) domain walls

are the $\{100\}_{pc}$ and $\{110\}_{pc}$ planes. The strain tensors as given in Table 1 have no common planes perpendicular to <100> or <110> because the strain refers to the cubic paraelastic system. For a convenient description of the domain states in an cartesian coordinate system, each strain tensor is added with the unit matrix **I** as

$$\mathbf{E}^{\mathbf{A}} + \mathbf{I} = \mathbf{D}^{\mathbf{A}} \tag{3}$$

such that $\mathbf{D}^{A}$ is affine mapping. To get the real orientations of the domains and of the domain walls in the ferroelastic phase, small rotations of the domains are necessary. We will call the rotated deformation matrices **M** orientation variants. The **M** matrices can be calculated by

$$\mathbf{M}^{\mathbf{A},\,\mathbf{i}} = \mathbf{R}^{\mathbf{i}}\mathbf{D}^{\mathbf{A}} \tag{4}$$

where $\mathbf{R}^{\,i}$ is a specific rotation matrix for domain state A.

The rotations $\mathbf{R}^{i}$ of the different $\mathbf{D}^{A}$ have to fulfill the following conditions.

1. The {100} and {110} planes of different rotated $\mathbf{D}^{A}$ have to be congruent.
2. Symmetry operations, that have been lost during the phase transition must act as generators between all $\mathbf{M}^{A,i}$ matrices of the orientation variants.
3. The misfit between the paraelastic cubic and the ferroelastic rhombohedral lattice should be as small as possible [11].

These conditions can only be satisfied with rotations of $\{100\}_{pc}$ planes onto $\{100\}_{c}$ with the additional restriction that the $<110>_{pc}$ directions will be rotated onto cartesian $<110>_{c}$ directions. The subscript *c* refers to the cartesian coordinate system of the paraelastic cubic phase. Table 3 lists the rotations with respect to certain compatible chevron twin-tilings.

Table 3. Rotations of the strain tensors in compatible twin tilings to obtain the orientation variants.

Possible domain walls in chevron twin tiling	Rotation of orientation states I - IV
(100), (110), $(1\overline{10})$	$(100)_{pc}$ & $[001]_{pc} \rightarrow (100)_{c}$ & $[001]_{c}$
(100), (101), $(10\bar{1})$	$(100)_{pc}$ & $[010]_{pc} \rightarrow (100)_{c}$ & $[010]_{c}$
(010), (110), $(\bar{1}10)$	$(010)_{pc}$ & $[001]_{pc} \rightarrow (010)_{c}$ & $[001]_{c}$
(010), (011), $(01\bar{1})$	$(010)_{pc}$ & $[100]_{pc} \rightarrow (010)_{c}$ & $[100]_{c}$
(001), (101), $(10\bar{1})$	$(001)_{pc}$ & $[010]_{pc} \rightarrow (001)_{c}$ & $[010]_{c}$
(001), (011), $(01\bar{1})$	$(001)_{pc}$ & $[010]_{pc} \rightarrow (001)_{c}$ & $[010]_{c}$

The connection of a certain twin-tiling with a certain rotation is necessary to maintain the symmetry connection between the paraelastic and the ferroelastic phase according to condition 2. Each chevron twin tiling contains only permissible domain walls, which can be calculated from equation (1). Therefore a $LaAlO_3$ crystal, which includes only permissible domain walls, consist at the most of 4 different orientation variants. As reported in [5] impermissible domain contacts, which violate the condition, that the two solutions of (1) are mutually exclusive, appear frequently in real crystals. There are altogether 6 × 4 = 24 different orientation variants.

The set of congruent symmetry elements within domains connected by permissible domain walls in a compatible patter as in Fig. 2 is called the stabilizer. The number of orientation variants corresponds to the group-subgroup index of the stabilizer in the point group of the paraelastic phase [12]. In the ferroelastic phase of $LaAlO_3$, the stabilizer of a domain pattern containing more than two domains includes the identity operation and the inversion. These symmetry elements generate the point group $\bar{1}$. Therefore, the number of orientation variants for $LaAlO_3$ is 24, as given by the order of the paraelastic point group (= 48) divided by the order of the stabilizer $\bar{1}$ (= 2).

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