Controlled modification of interfacial strain and twinning in $YBa_2Cu_3O_{7-\delta}$ films on vicinal $SrTiO_3$ (001)

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(Received 30 June 1997)

The structure of twinning in $YBa_2Cu_3O_{7-\delta}$ thin films grown on vicinal $SrTiO_3$ (001) by pulsed laser deposition has been studied as a function of the vicinal miscut angle by x-ray diffraction using the two-dimensional q-scan technique with a four circle diffractometer. Our results reveal a strong correlation between the miscut of a $SrTiO_3$ (001) substrate and the strain occurring at the interface between substrate and film. Depending on the proper choice of miscut orientation and angle, one of the two twin systems can be suppressed and ≈70% untwinned growth can be achieved. [S0163-1829(98)00406-8]

Single crystals and epitaxial films of $YBa_2Cu_3O_{7-\delta}$ (YBCO) are typically twinned, with twin boundaries aligned along [110] and $[\bar{1}10]$. Furthermore, the electrical transport properties, as well as flux pinning in epitaxial YBCO films are extremely sensitive to the underlying microstructure. Specifically, it has been found that the twin boundaries cause correlated pinning as well as a channeling of vortices along twin planes.[1–5] Thus, there is considerable interest in controlling the twin structure. It has been reported that almost twin-free YBCO films can be grown on orthorhombic $NdGaO_3$ (001).[6] However, a systematic study of the interrelationship between the structure and morphology of the substrate surface and the epitaxial orientation of the film is still missing.

Recently, Haage and co-workers[7] have generated periodic step structures of clean and YBCO-covered vicinal $SrTiO_3$ (001) surfaces. The resultant film microstructure leads to an in-plane anisotropy of the transport properties and flux pinning in the superconducting material.[8] Here we report the observation of anisotropic strain at the substrate/film interface in YBCO films on vicinal $SrTiO_3$ (001). Furthermore, we demonstrate how to control the formation of twin boundaries via film growth on $SrTiO_3$ (001) with a different miscut.

The $SrTiO_3$ (001) substrates were cut and polished 0.4° to 10° towards [010] (and [110]) off (001). UHV annealing at 930 °C over 2 h sufficed to generate clean and stepped surfaces, as described in detail by Haage and co-workers.[7] $YBa_2Cu_3O_7$ films were grown by pulsed laser deposition with a thickness of 24 to 240 nm.

The x-ray-diffraction measurements were performed on a STOE four-circle diffractometer with graphite monochromatized Co $K\alpha$ radiation ($\lambda = 1.790200$ Å). The tube operated at 40 kV and 30 mA. By a divergence of 0.3° the diameter of the spot on the sample was approximately 1.2 mm. The distance between the detector and the sample was 250 mm while the horizontal and vertical detector slits were 1.5 mm.

Two-dimensional scans in reciprocal space (q scans) were performed to investigate the splitting of the YBCO reflections.[9] Within layers of constant/selected ranges of h and k were analyzed. In the following the indices h and k are given in units of the reciprocal $SrTiO_3$ lattice.

The twinning of YBCO occurs at $(\bar{1}10)$ and (110) planes of intergrowth. If the initial stage of growth of the film is epitaxial, the $[\bar{1}10]$ and [110] directions of YBCO are strictly parallel to the $[\bar{1}10]$ and [110] directions of a $SrTiO_3$ (001) substrate. Therefore, YBCO grown on well-oriented $SrTiO_3$ (001) exhibits a regular splitting of the reflections, reflecting the orthorhombic symmetry of the individual domains.[10]

The lattice mismatch between the cubic $SrTiO_3$ substrate ($a = 3.905$ Å) and the orthorhombic YBCO layer ($a = 3.8227$ Å, $b = 3.8872$ Å, $c = 11.6802$ Å) causes stress and strain at the interface. On well-oriented $SrTiO_3$ (001) substrates the in-plane strain is isotropic and does not affect the twinning of the film. Carefully annealed vicinal $SrTiO_3$ (001) surfaces with a miscut angle of Θ towards [010] exhibit straight step edges aligned along [100].[7] Most steps are 0.39 nm high (0.39 nm = unit cell height of $SrTiO_3$). With Θ ranging from ≈0.4° to ≈10° we observed 56 to 2.3 nm-wide $SrTiO_3$ (001) terraces, respectively.

Due to the step structure on the vicinal surfaces of the used substrates, the terminating layer of $SrTiO_3$ can be strained anisotropically by the YBCO film. The anisotropic strain at the interface, which depends on the angle and direction of the miscut, strongly affects the twinning geometry and the relative intensities of the split YBCO reflections.

The twinning of a 60 nm-thick YBCO film grown on 10° miscut $SrTiO_3$ (001) (towards [010]) is shown in Fig. 1. The subscripts $S_1- \ldots S_2+$ in this figure describe the four different suborientations according to Wadhawan.[11] The reflections S_1+ and S_2- correspond to the twinning system that is attached to the substrate surface towards [110] (henceforth termed $[S_1+, S_2-]$). The two lines and the two dotted lines in the figure mark the k and h values, respectively, for a regularly twinned YBCO film on a well-oriented $SrTiO_3$ (001) substrate. Due to the miscut of the substrate, the twinning systems $[S_1+, S_2-]$ and $[S_1-, S_2+]$ are rotated by −0.095° and 0.095°, with respect to the drawn lines. There-

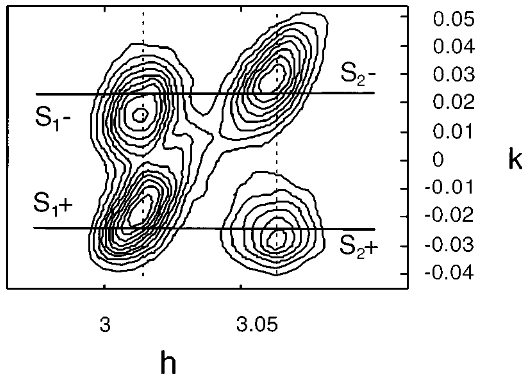

FIG. 1. q scan of the split YBCO 302 reflection of a 60 nm-thick YBCO film grown on $SrTiO_3$ (001) with a miscut of 10° towards [010] and 0.28° towards [110]. The indices h and k are given in units of the reciprocal $SrTiO_3$ lattice. The two solid lines and the two dotted lines mark the k and the h values, respectively, for a regularly twinned YBCO film on a well-oriented $SrTiO_3$ (001) substrate.

fore the angle between the two twinning systems with twin boundaries along [110] and $[\bar{1}10]$ is 90.19°. This finding suggests that the terraces of the substrate are tetragonally distorted since they can be strained more easily perpendicular than parallel to the step edges (i.e., towards [010]). Consequently, the angle ζ between the two directions of intergrowth [110] and $[\bar{1}10]$ exceeds 90° (Fig. 2). As the epitaxy only depends on the surface structure of the substrate the two twinning systems of the YBCO film are rotated, with respect to each other and the substrate.

Assuming that all other torsional forces in thin films can be neglected, the relative strain $\Delta\varepsilon_{[01]}=\varepsilon_{[01]}-\varepsilon_{[10]}$ between [010] and [100] of the top layer of the substrate surface can be calculated from the angle ζ between the two twinning systems of the film. We find $\Delta\varepsilon_{[01]}=1-\tan(\zeta/2)$. Therefore the measured angle ζ=90.19° in Fig. 1 leads to an anisotropic strain of $\Delta\varepsilon_{[01]}=-0.33\%$. The surface of the vicinal $SrTiO_3$ (001) is more compressed by the YBCO film towards the direction of miscut [010]. To the best of our knowledge this is the first reported investigation of strain at the interface between a crystal and an epitaxial layer. Investigating the strain by the torsion angle ζ is most conveniently realized by the here presented q-scan technique employing a conventional x-ray tube.

Moreover, a preferred orientation within the two twin systems due to the anisotropic strain at the interface is found. The intensity of the reflections S_1- and S_1+, which represent the two film orientations with the b axis parallel to [100] (i.e., along the step edges), are 1.78 times larger than S_2+ and S_2-, respectively (Fig. 1). The shorter a axis of the YBCO film is preferably oriented along the [010] direction of the $SrTiO_3$ substrate (i.e., perpendicular to the step edges). In Fig. 3. the influence of the anisotropic strain at the interface, measured by the torsion angle ζ between the two twin-

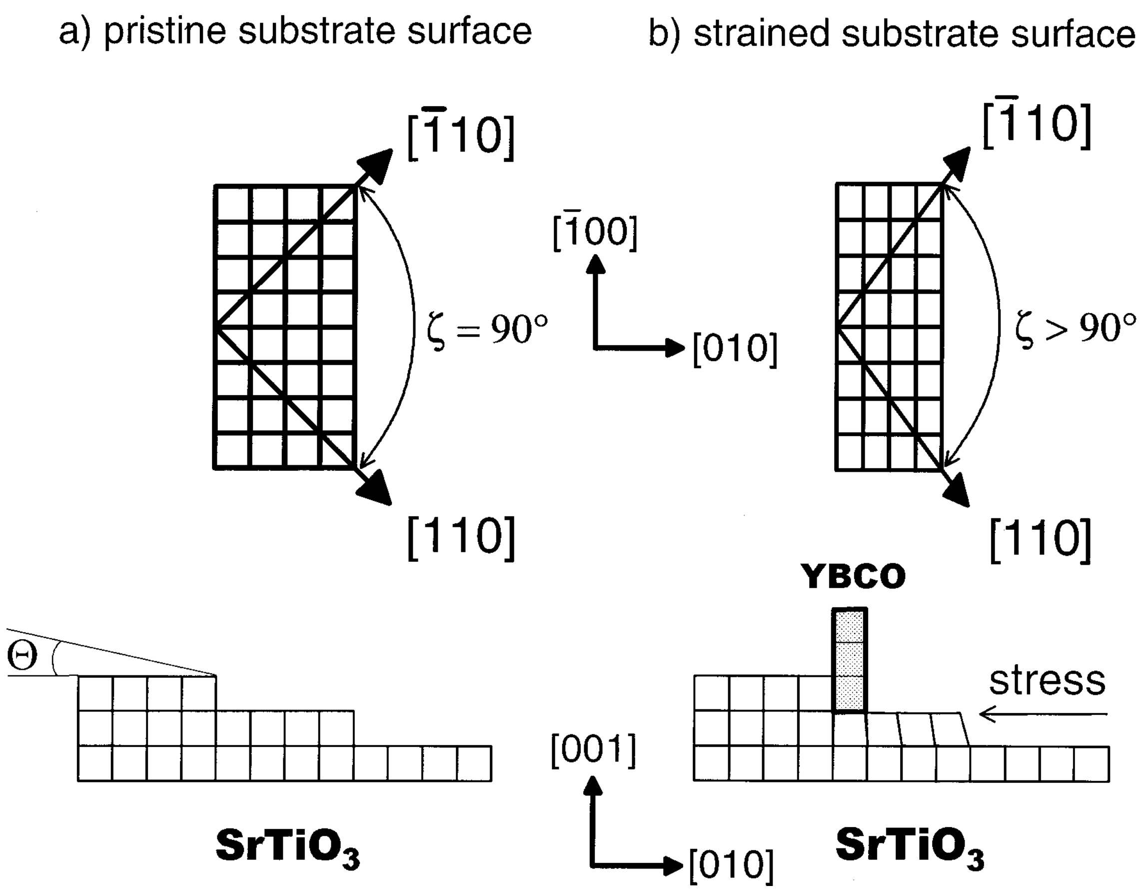

FIG. 2. (a) Terrace structure of a pristine vicinal $SrTiO_3$ (001) surface with a miscut of Θ° towards [010]. The angle ζ between [110] and $[\bar{1}10]$ is 90°. (b) Terrace structure of a YBCO-covered vicinal $SrTiO_3$ (001) surface. Due to the lattice mismatch, the substrate surface is tetragonally distorted and the angle ζ exceeds 90°.

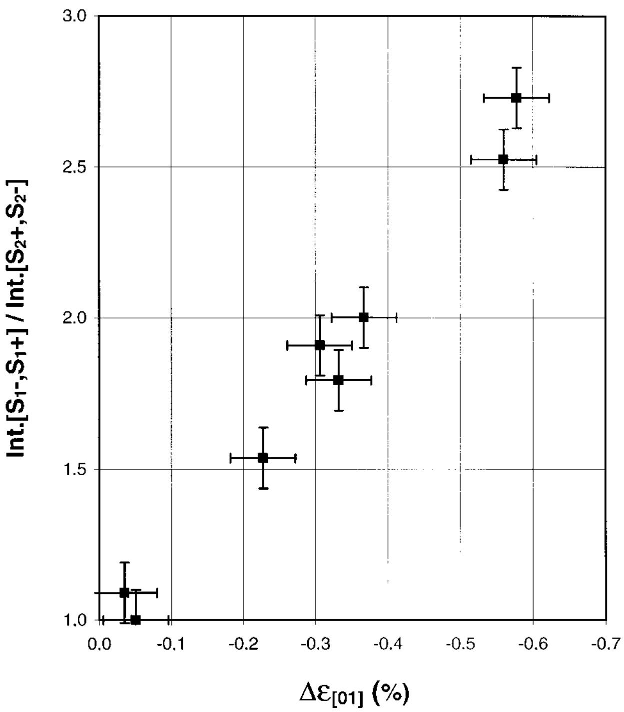

FIG. 3. Detwinning of the YBCO film as a result of the anisotropic strain $\Delta\varepsilon_{[01]}$ at the substrate surface. The values at the y axis represent the sum of the determined peak intensities S_1- and S_1+, normalized to the peak intensities S_2- and S_2+ (see also Fig. 1).

ning systems, on the preference of the YBCO orientation *within* a twinning system is shown for films grown on vicinal $SrTiO_3$ surfaces with different tilt angle. Increased vicinal angles are associated with a smaller terrace width. Since the surface lattice can be strained more easily towards [010], reducing the terrace width enables a tailoring of the anisotropic strain.

For comparison, the twinning of YBCO grown on tetragonal $NdGaO_3$ (100) substrate with the lattice constants a, b $=3.863$ Å and $c=3.854$ Å reveals an angle of 90.13° between the two directions of intergrowth [011] and $[0\bar{1}1]$ due to the slight orthorhombicity of 0.23%. The resulting preference of the two S_1 orientations (Int.$[S_1+,S_1-]$/Int.$[S_2-,S_2+]=1.55$) (Ref. 12) is in good agreement with the strain-induced preference of the S_1 system for our $SrTiO_3$ substrate with $\Delta\varepsilon_{[01]}=-0.23\%$ (Fig. 3). By increasing the film thickness up to 1200 Å the rotation between the two twinning systems and consequently the anisotropic strain at the vicinal $SrTiO_3$ (001) surface was enhanced. The resulting preference of the S_1 system for these samples is significantly increased compared with the S_1 preference for the $NdGaO_3$ substrate (Fig. 3).

Due to the lattice mismatch at the interface, the film is also under tensile stress. A reduced lattice dilation of the film towards the [010] direction of the substrate was observed for all four suborientations. This confirms our suggestion that the substrate surface is tetragonally distorted. By increasing the film thickness the lattice parameters of the film are reduced and converge to the stress-free lattice constants.

In Fig. 4, the twinning of a 60 nm-thick YBCO film on $SrTiO_3$ with a miscut of 2.6° towards [110] is shown. It is

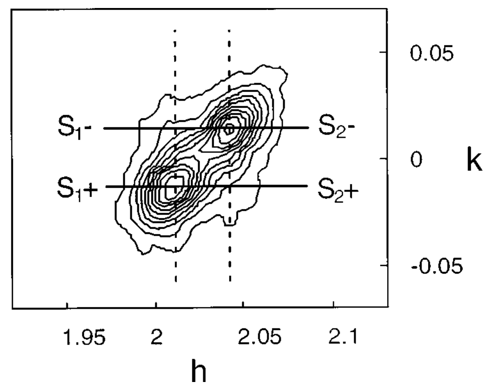

FIG. 4. q scan of the split YBCO 205 reflection of a 60 nm-thick YBCO film grown on $SrTiO_3$ (001) with a miscut of 2.6° towards [110]. The indices h and k are given in units of the reciprocal $SrTiO_3$ lattice. The two solid lines and the two dotted lines mark the k and the h values, respectively, for a regularly twinned YBCO film on a well-oriented $SrTiO_3$ (001) substrate.

obvious that the twinning system $[S_1+,S_2-]$ is strongly preferred. Since the strain towards [110] does not change the angle between [110] and $[1\bar{1}0]$, the suborientations of the YBCO film are not rotated ($\zeta=90°$).

Vicinal $SrTiO_3$ (001) surfaces with a miscut towards [110] exhibit a regular kink structure.[13] Our findings suggest that the $SrTiO_3$ terraces can be strained more easily along [110] than along $[1\bar{1}0]$. Therefore, the lattice mismatch along [110] is significantly reduced compared to the $[1\bar{1}0]$ direction. The twinning system $[S_1+,S_2-]$, which is attached to the substrate along [110], is therefore preferred. Even substrates with a miscut up to 10° towards [010] and a small additional miscut towards [110] reveal a strong preference of one twinning system. In Fig. 1. the preference of the

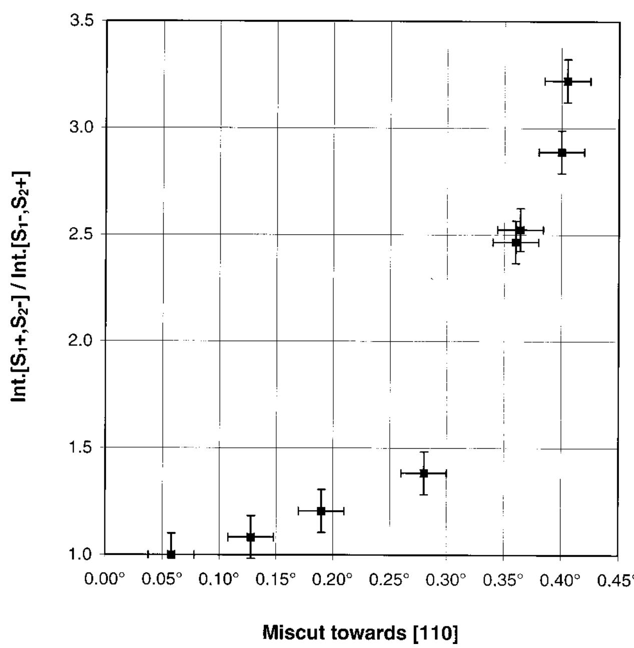

FIG. 5. Preference of the $[S_1+,S_2-]$ twinning system of the YBCO film as a result of the $SrTiO_3$ (001) miscut towards [110]. The values at the y axis represent the sum of the determined peak intensities S_1+ and S_2-, normalized to the peak intensities S_1- and S_2+ (see also Fig. 4).

twinning system $[S_1+,S_2-]$ aligned in [110] is due to an additional miscut of 0.28° towards [110].

The anisotropy of the strain occurring at the interface can be enhanced by increasing the angle of the miscut. In Fig. 5, the influence of the miscut towards [110] on the preference of the $[S_1+,S_2-]$ twinning system is shown for different samples. It is evident that even a small (additional) miscut towards [110] enables an anisotropic strain at the substrate surface, leading to a preference of one twinning system. Samples with a larger miscut towards [110] reveal a preference of ≈100% (Fig. 4). The preference of the suborientations is more strongly influenced by a miscut towards [110] than towards [010]. This finding demonstrates that the film is considerably stronger attached to the substrate towards $\langle hh0\rangle$ than towards $\langle 0k0\rangle$.

Furthermore, a reduced lattice expansion of the preferred twinning system due to the anisotropically strained interface was observed. The influence on the transport properties will be discussed in a forthcoming publication.[14]

In conclusion, we have presented a method to investigate the strain at the interface between vicinal $SrTiO_3$ (001) substrate surfaces and $YBa_2Cu_3O_7$ thin films with x-ray diffraction. Anisotropic strain created by the substrate surface leads to a ≈100% suppression of one twin system or/and a ≈70% untwinning of the film, i.e., 70% suppression within a twin system, depending on the proper choice of miscut orientation and angle.

This work was supported by the German BMBF (BMFT) under Contract No. 13N5840.

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