Bulk-sensitive Imaging of Twin Domains in La$_{2-x}$Sr$_x$CuO$_4$ under Uniaxial Pressure

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(Dated: 6 October 2018)

We report our study of twin domains in La$_{2-x}$Sr$_x$CuO$_4$ under uniaxial pressure. Using bulk-sensitive x-ray microdiffraction in Laue geometry, we image the distribution of twin domains at room temperature. When compressive uniaxial pressure is applied along one of the in-plane crystallographic axes, the domain population changes dramatically. We observe that the twin domain with shorter lattice parameter along the direction of pressure is unstable under compression, and disappears completely with only moderate pressure. On the other hand, application of tensile pressure changes the domain structure only slightly, demonstrating the asymmetric response of the sample to uniaxial pressure. Our observations suggest that a crystal’s response to uniaxial pressure is complex and could deviate easily from the linear-response regime.

Understanding of stress and strain has been an integral part of materials science and engineering. Mechanical properties of bulk engineering materials are influenced significantly by the residual stress, while epitaxial strain plays an important role in determining structural and electronic properties of many thin film materials. Unlike such naturally occurring, unavoidable stress/strain, a deliberate application of uniaxial stress/strain, a deliberate application of uniaxial pressure can also directly modify electronic properties in graphene as well as the superconducting transition temperature in Sr$_2$RuO$_4$. Detwinned cuprate superconductors through the tetragonal-orthorhombic structural transition with applied uniaxial pressure removes one of the twin domains, allowing anisotropic transport properties in the FeAs plane to be studied. Uniaxial pressure can also directly modify electronic properties in graphene as well as the superconducting transition temperature in Sr$_2$RuO$_4$. Detwinned cuprate superconductors through the tetragonal-orthorhombic structural transition with applied uniaxial pressure removes one of the twin domains, allowing anisotropic transport properties in the FeAs plane to be studied. Uniaxial pressure can also directly modify electronic properties in graphene as well as the superconducting transition temperature in Sr$_2$RuO$_4$. Detwinned cuprate superconductors through the tetragonal-orthorhombic structural transition with applied uniaxial pressure removes one of the twin domains, allowing anisotropic transport properties in the FeAs plane to be studied. Uniaxial pressure can also directly modify electronic properties in graphene as well as the superconducting transition temperature in Sr$_2$RuO$_4$. Detwinned cuprate superconductors through the tetragonal-orthorhombic structural transition with applied uniaxial pressure removes one of the twin domains, allowing anisotropic transport properties in the FeAs plane to be studied. Uniaxial pressure can also directly modify electronic properties in graphene as well as the superconducting transition temperature in Sr$_2$RuO$_4$. Detwinned cuprate superconductors through the tetragonal-orthorhombic structural transition with applied uniaxial pressure.

A superconducting La$_{1.93}$Sr$_{0.07}$CuO$_4$ (LSCO x=0.07) crystal was used in our study, which possesses a layered perovskite structure with an orthorhombic to tetragonal structural phase transition at a temperature $T_s \approx 400$ K. In the high temperature regime, the LSCO sample is in tetragonal structure with the $I4/mmm$ symmetry. Below $T_s$, the crystal symmetry becomes $Bma_{h}$, and...
FIG. 2. (a) Partial view of the full diffraction pattern under no strain, showing five pairs of diffraction peaks. Also shown are the close-up views at -20 V (compressive strain), 0 V (no strain), and +30 V (tensile strain). (b) The reciprocal space perpendicular to the c-axis for the two twin domains (different colored axes). The filled circles are the Bragg peak positions for (200) and (020) peaks for the two domains. The evolution of the peak intensity with compressive strain is shown in the middle. (c) Schematic real-space picture illustrating the response of CuO$_2$ planes in LSCO under compressive strain. The buckled rectangles represent CuO$_6$ octahedra. Note the elongation along the compression direction.
ence sample in the same diffraction setup. Although the angular resolution was not very high, the Laue geometry and a perfect crystal sample allow us to refine sample strain (see Supplementary Material). The sample strain along the uniaxial pressure direction was determined to be about $3 \times 10^{-4}$ when $+25$ V was applied to the piezo stack. Since the bulk modulus of silicon (100 GPa) is not too different from that of LSCO (130 GPa), one can then estimate the uniaxial pressure being applied in our LSCO experiment to be about 1 MPa/V. However, due to the large uncertainty in this estimate, we quote the voltage applied to the piezo stack in this paper without converting to pressure. A negative voltage $v$ represents a voltage $|v|$ applied to the middle piezo (compressive strain), while a positive voltage represents a voltage applied to the outer piezos (tensile strain). Throughout this paper, we use the convention that a (+) sign means tensile strain and a (-) sign means compressive strain.

In Fig. 2 (a), we show a portion of the diffraction pattern of LSCO taken at the center of the sample under no stress. Five pairs of diffraction peaks due to twin domains can be seen. (Full diffraction images can be found in Supplementary Material.) We also show close-up images of a select pair of peaks under various strains: $-20$ V, $0$ V, and $+30$ V respectively. Before application of pressure (0 V), both domains are present and a pair of peaks are observed. At $-20$ V, the bottom peak vanishes and the sample becomes single domain at this location. At $+30$ V, the bottom peak intensifies slightly and there seems to be some change in the domain population.

The Laue scattering geometry in which the incoming x-ray beam is parallel to the c-axis is advantageous for finding out which domain each peak belongs. We show the schematic reciprocal space projection on the detector plane in Fig. 2(b). Here the reciprocal space coordinate corresponding to the $a(b)$-axis oriented horizontally is shown in a blue solid line (red dot-dash line). The rotation angle between the two coordinate systems is exaggerated in this figure. The pair of peaks along these axes correspond to the $(200)/(020)$ pair coming from the two twin domains. The diffraction data obtained at 0 V, $-5$ V, and $-20$ V for each pair are shown in the center panels, allowing one to follow the peaks and see which of them disappears when compressed. Between the two peaks on the vertical axes, one can clearly see that the peak on the right disappears, while the bottom peak disappears on the horizontal axes. Both disappearing peaks belong to the same twin domain (red dot-dash), as expected. But what is surprising is the direction of applied compressive pressure (dashed arrows). Our results indicate that the compressive strain stabilizes the twin domain with the longer lattice parameter (i.e., $b$) along the compression direction. That is, the twinned crystal of LSCO displays a negative compressibility. This will be discussed further below.

The domain populations appeared to respond to compressive and tensile strain very differently. To characterize this difference, the pressure (voltage) dependence of peak intensity ratio was studied. For a given voltage, the ratio of the integrated intensity of the bottom peak to the top peak was calculated for each of the five pairs of diffraction peaks in Fig. 2(a). This number was then averaged over the five pairs and plotted against the voltage as shown in Fig. 3. Now the asymmetry is made apparent. A $-20$ V compressive strain easily eliminates one domain, while both domains persist up to 30 V tensile strain. In addition, substantial hysteresis is observed in the case of tensile strain. We note that the piezo stack itself shows a mild hysteretic behavior, but in the opposite direction. Therefore, we believe that the observed hysteresis behavior is a characteristic of the sample under tensile strain.

In order to study spatial distribution of twin domains, we scanned the sample in the $ab$-plane over a 240 $\mu$m x 240$\mu$m area near the center of the sample. The step size used roughly matches the beam size (3 $\mu$m x 3 $\mu$m). At each point the same diffraction pattern as shown in Fig. 2 was obtained, and the averaged ratio of the 5 pairs was obtained in the same manner as before. A pseudocolor map presented in Fig. 4 was then generated using these averaged ratios as intensities, giving a characterization of the domain population distribution in the sample. The stripy pattern in Fig. 4 arises from the twin domains. We observe that the domain walls run along the (110) direction, and their separation is on the order of 10 microns. This is consistent with earlier observations from optical or electron microscopy. The twin domain structure observed after the removal of uniaxial pressure seems to be identical to the original domain structure before the application of uniaxial pressure.

In Fig. 4 (f), we plot the ratio along a line perpendicular to the domain walls in the 30 V map and its Fourier
FIG. 4. Domain population ratio map at various voltages. (a)-(e) Domain population ratio map for −20 V, −5 V, 0 V, 20 V, and 30 V. The scale of the color bar is logarithmic. (f) A plot of the ratio along a cut perpendicular to the striping pattern in the 30 V map. Inset: An indication of the location of the cut and the Fourier transform of the cut in the wavelength domain.

transform. The absence of any particular periodicity confirms the random nature of the domain size, which are presumably pinned by lattice defects. If the domains are much smaller than the spatial resolution determined by the beam size, then we expect a more homogeneous intensity distribution due to spatial averaging. The fact that we see these variations suggests that the domain size is at least comparable to the beam size. In addition, the presence of these patterns also mean that the domains are fairly well correlated along the c-direction. Significant variation in the domain population along the c direction would result in a featureless map in this transmission geometry as an average is taken when the beam traverses through the sample of 100 µm thickness. We also note that the intensity ratio variation (e.g. for 0 V data) is much smaller than what is expected from an ideal domain distribution (zero to infinity in this ideal case). This could be due to the microtwins found inside larger twin domains as reported in Ref. 16. To ensure that the observed pattern is an intrinsic sample property, we repeated the measurements with the reverse experimental geometry. When the strain device was rotated by 180 degrees about the vertical axis (the a(b) axis in Fig. 1) so that the sample front is facing the beam, the observed domain pattern also flips as expected, confirming the intrinsic nature of the observed pattern. (Supplementary Material.)

To summarize, we show that application of compressive uniaxial pressure along the orthorhombic a or b axis of La$_{1.93}$Sr$_{0.07}$CuO$_4$ can completely eliminate the twin domain with the a-direction along the uniaxial direction. This is a very surprising discovery by virtue of the fact that a is smaller than b. So why does the crystal prefer to elongate under uniaxial compression? Although a quantitative theory is lacking, we can gain some qualitative insights by examining why La$_2$CuO$_4$ becomes orthorhombic below 450 K in the first place. This is often attributed to bond-length mismatch. The Cu-O equilibrium bond length is too large compared to the equilibrium bond length of La-O, placing the CuO$_2$ layer under compressive stress and the LaO layer under tension even in the absence of applied pressure. Buckling of the CuO$_6$ octahedra addresses this problem because tilting of a CuO$_6$ octahedron brings the apical oxygen closer to La without shortening the Cu-O bond length. Therefore, apparently, application of compressive stress to (already stressed) Cu-O bonds along the a-axis causes them to buckle and become the b-axis (See Fig. 2(c) for a schematic illustration). The buckling also distorts the CuO$_6$ octahedra, which causes the b-axis to have a larger lattice parameter. In fact, rotation of rigid structural units plays an important role in many materials exhibiting negative compressibility. 24, 25 It should be noted that the layered structure of La$_{2-x}$Sr$_x$CuO$_4$ makes this a particularly simple system to study physics of octahedral tilting and uniaxial strain, unlike three-dimensional
structures such as perovskites, in which octahedral tilting and rotation in response to pressure can be quite complicated.

Our findings have several interesting implications for future studies. We demonstrated the power of x-ray Laue microdiffraction as a tool for studying strain in materials. In particular, the combination of spatial resolution and bulk-sensitivity allows one to study the response of twin domains in detail. Further investigation of various negative compressibility materials could be useful for elucidating non-linear response of materials under uniaxial pressure.

SUPPLEMENTARY MATERIAL

See Supplementary Material for the strain refinement of the silicon reference sample, the reverse experimental geometry data, and the full diffraction image of the peaks shown in Fig. 2(a).

ACKNOWLEDGMENTS

Work at the University of Toronto was supported by the Natural Science and Engineering Research Council (NSERC) of Canada through Discovery Grant (RGPIN-2014-06071). Canadian Foundation for Innovation, and Ontario Innovation Trust. Research performed at the Canadian Light Source is supported by the Canada Foundation for Innovation, NSERC, the University of Saskatchewan, the Government of Saskatchewan, Western Economic Diversification Canada, the National Research Council Canada, and the Canadian Institute of Health Research. X. Y. Z. acknowledges the receipt of support from the NSERC Undergraduate Student Research Award.

1We use the term pressure in our experiment, since both stress and strain are second-rank tensor quantities, and thus not easy to define uniaxial direction.

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