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Reversible shift in the superconducting transition for La$_{1.85}$Sr$_{0.15}$CuO$_4$ and BaFe$_{1.8}$Co$_{0.2}$As$_2$ using piezoelectric substrates

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Abstract. The use of piezoelectric substrates enables dynamic observation of the strain-dependent properties of functional materials. Based on studies with La$_{1.85}$Sr$_{0.15}$CuO$_4$ (LSCO), we extended this approach to the iron arsenic superconductors represented by BaFe$_{2-x}$Co$_x$As$_2$ to investigate strain-driven changes in detail. We demonstrate that epitaxial thin films can be prepared on (001) Pb(Mg$_{1/3}$Nb$_{2/3}$)$_{0.72}$Ti$_{0.28}$O$_3$ substrates using pulsed laser deposition. The structural and electric properties of grown films were characterized in detail. A reversible shift of the superconducting transition of 0.4 K for LSCO and 0.2 K for BaFe$_{1.8}$Co$_{0.2}$As$_2$ was observed on applying biaxial strains of 0.022 and 0.017%, respectively.

The application of pressure has a significant influence on the physical properties of functional materials. Detailed experiments are required to enable a deeper understanding of these materials in physics, especially the sensitive interplay between structural parameters such as bonding length or angle and electronic properties. Existing studies on bulk materials, predominantly cuprate high-temperature superconductors, using hydrostatic pressure demonstrate that compressive pressure increases the superconducting transition temperature, T_c, for most materials. Recent studies also reveal a strong influence of pressure on superconductivity for iron-based superconductors, resulting in a pressure-dependent superconducting dome in the electronic phase diagram similar to doping [1, 2]. It should be noted that this shift in T_c is

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highly anisotropic regarding strain along different crystallographic axes. For some oxides such as $La_{2-x}Sr_xCuO_4$, it is known that the resulting effect is partially neutralized for hydrostatic pressure [3]–[6].

Therefore, in the last decade the application of biaxial strain also attracted increasing attention, especially for the model system $La_{2-x}Sr_xCuO_4$ [7, 8]. Similarly, recent studies on biaxial strained iron-based superconductors such as $BaFe_{1.8}Co_{0.2}As_2$ (Ba-122) and $FeSe_{0.5}Te_{0.5}$ also revealed that compressive biaxial strain enhances T_c [9, 10]. Typically, epitaxial thin films are prepared on various single crystalline substrates with a different lattice mismatch between substrate and film inducing a biaxial tensile or compressive strain. However, this approach is often restricted to very thin films because of a limited layer thickness for coherent strained growth. A large misfit typically leads to partial relaxation of the lattice and, therefore, to the implementation of lattice defects. In this case, it is difficult to correlate the applied strain with superconducting properties directly, because the preparation conditions and the resulting microstructure may severely affect the latter.

An alternative approach to static pressure experiments is the preparation of super-conducting films on single crystalline piezoelectric substrates. Using the inverse piezoelectric effect, the applied strain can be changed continuously and reversibly by an electric field. This approach offers a unique opportunity to investigate the strain-dependent properties on one and the same sample, as already shown for ferromagnetic oxides [11]–[14]. We recently reported on the epitaxial growth of superconducting $YBa_2Cu_3O_{7-\delta}$ and $La_{1.85}Sr_{0.15}CuO_4$ (LSCO) thin films on pseudocubic (001) $Pb(Mg_{1/3}Nb_{2/3})_{0.72}Ti_{0.28}O_3$ (PMN-PT) substrates [15, 16]. In this paper, we extend this approach to Ba-122 thin films and report on a reversible shift in T_c with applied strain.

For sample preparation on PMN-PT we used a standard pulsed laser deposition (PLD) setup equipped with a Lambda Physiks LPX 305 KrF laser and stoichiometric targets. To reduce the lattice mismatch between PMN-PT ($a = 4.02$ Å) and the superconducting film, we deposited smooth 20 nm thick buffer layers of either $SrTiO_3$ (STO) ($a = 3.905$ Å) or $CaTiO_3$ (CTO) ($a = 3.82$ Å) [17]. The buffer layers as well as the 300 nm thick LSCO films are prepared in 0.3 mbar oxygen atmosphere at substrate temperatures of 650–$700\,^\circ$C using off-axis deposition [13, 18]. Films prepared under this condition typically exhibit a very smooth surface and droplet-free growth [18]. Subsequently, the films were cooled down in 0.4 bar oxygen atmosphere. A detailed description of LSCO preparation as well as structural analysis can be found in our previous paper [16]. A scheme of the film architecture is given in figure 1.

For the preparation of Ba-122, we used STO-buffered PMN-PT prepared by off-axis PLD as described above. Subsequently, the substrate was transferred to an ultra-high vacuum system with a base pressure of 10^{-9} mbar where the Ba-122 was deposited at $650\,^\circ$C using on-axis PLD. A detailed description of the film preparation can be found in Iida *et al* [10, 19].

The superconducting properties were characterized in a Quantum Design physical properties measurement system (PPMS) using a four-probe technique. For the evaluation of the transition temperature, a 50% resistance criterion is used. To confirm epitaxial growth and to study the structural properties, standard x-ray diffraction (XRD) in Bragg–Brentano geometry, pole figure measurements and reciprocal space mapping (RSM) were performed using a Phillips XPert MRD Diffractometer with Cu K_α radiation. X-ray reflectivity was used to determine the layer thickness and the roughness of the buffer layers.

We achieved a *c*-axis oriented growth and cube on cube epitaxy for both buffer layer and LSCO [16]. Also for Ba-122 the pole figure of the (103) peak, given in figure 2(a), proves
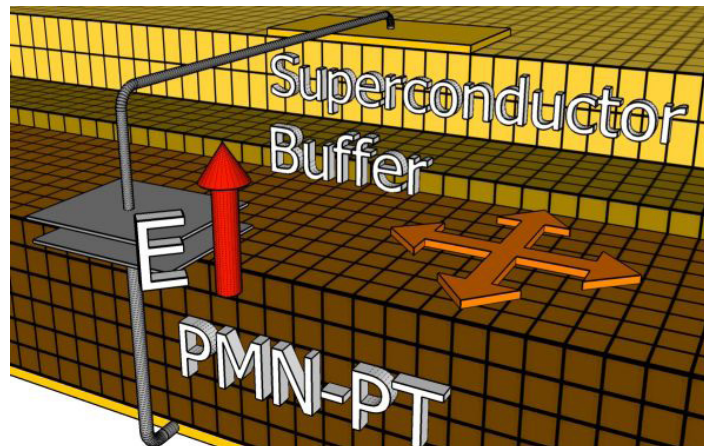
Figure 1. Schematic film architecture. The bottom contacts are sputtered NiCr/Au, whereas the top electrodes are deposited gold.
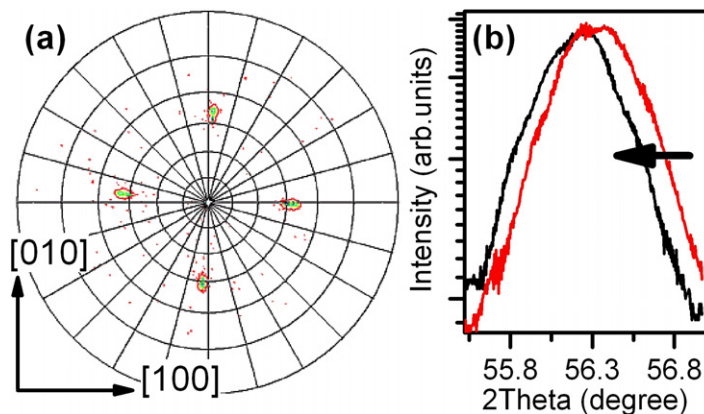

Figure 2. (a) Pole figure of the Ba-122 (103) reflection. The intensity is scaled quadratic, where maximum intensity corresponds to 650 cps; (b) the Ba-122 (008) peak is shifted to lower angles when compressive in-plane strain is applied.

perfect cube on cube epitaxy without any misorientation because the Ba-122 peaks are oriented parallel to the substrate [100] directions. The superconducting transition of LSCO at 17.5 K on CTO-buffered PMN-PT is slightly smaller compared to 18.5 K on STO-buffered PMN-PT. However, the CTO-buffered system was used for further investigations due to reduced affinity to crack during dynamic strain measurements.

In the case of Ba-122 the STO-buffered films exhibit a T_c of 14 K, which is significantly reduced compared to 23 K for films prepared on bare STO [10]. Part of this reduction is attributed to the poorer crystalline quality of the PMN-PT substrate compared to STO and the much larger transition width as we use a 50% criterion.

In the first step, it is necessary to ensure the transfer of strain into the superconducting layer. Detailed investigations by Bilani *et al* showed that the strain is transferred from the PMN-PT to the STO buffer [17]. We performed additional high-resolution XRD and RSM to verify strain transfer to the superconducting layer. An example is given in figure 2(b) for the Ba-122 (008) peak without and with a 16.6 kV cm^{-1} applied field.
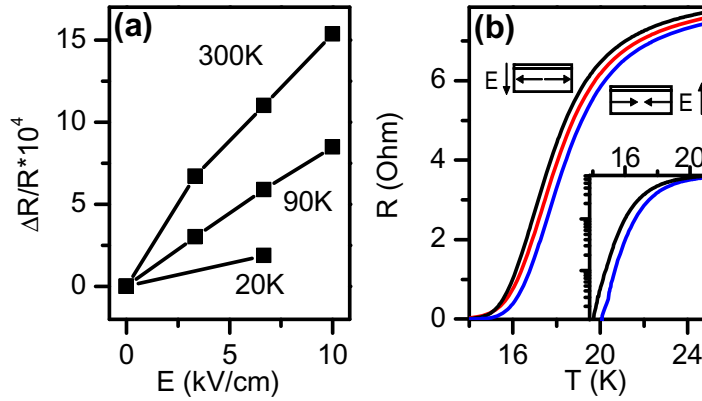
Figure 3. (a) Relative change of the resistivity of a platinum strain gauge in dependence on the field for three different temperatures; (b) shift of transition temperature with applied field for LSCO for $E = -10\,\mathrm{kV\,cm^{-1}}$ (black), $E = 0\,\mathrm{kV\,cm^{-1}}$ (red) and $E = 10\,\mathrm{kV\,cm^{-1}}$ (blue). The inset shows the logarithmic scaled resistivity.

The change of the lattice parameters in PMN-PT single crystals at room temperature in dependence on the applied electric field is well investigated [20]; however, there are no data available for lower temperatures. Nevertheless, knowledge of the low-temperature behavior is essential to correlate the strain with the change in the superconducting properties.

To gauge the magnitude of strain at lower temperatures, we deposited a thin meander-shaped platinum wire at room temperature directly on CTO-buffered PMN-PT. The resistivity of the wire correlates to the biaxial strain because of a change in wire geometry. The change in relative resistivity with applied electric field is given in figure 3(a) for three different temperatures. These data reveal a strong reduction of the strain at constant electric field with decreasing temperature. Compared to room temperature ($\epsilon_a = 0.12\%$ at $10\,\mathrm{kV\,cm^{-1}}$), we achieve half the value at 90 K and less than 20% at 20 K. The biaxial in-plane strain, ϵ_a, is defined as $(a_0 - a_\mathrm{strained})/a_0$, where a_0 is the unstrained in-plane lattice parameter.

To check the suitability of our approach, we used the well-known model system LSCO. Applying an electric field of $E = 10\,\mathrm{kV\,cm^{-1}}$ at 20 K, which corresponds to $\epsilon_a = 0.022\%$, we achieved a reversible shift of the superconducting transition temperature of 0.4 K (figure 3(b)). We compared this shift with available literature data using a simple equation of the strain-dependent T_c for an orthorhombic unit cell, where $T_c(0)$ denotes the superconducting transition temperature of the unstrained film:

$$T_c = T_c(0) + \frac{\delta T_c}{\delta \epsilon_a}\epsilon_a + \frac{\delta T_c}{\delta \epsilon_b}\epsilon_b + \frac{\delta T_c}{\delta \epsilon_c}\epsilon_c. \tag{1}$$

The values for the derivatives are well investigated for LSCO [4]. Because of the biaxial strain, ϵ_a equals ϵ_b. Taking the correlation of ϵ_c and ϵ_a from statically strained LSCO films [8], we can replace ϵ_c in the out-of-plane term and finally summarize equation (1) as

$$T_c = T_c(0) + \beta \epsilon_a, \tag{2}$$

where $\beta = 2000$ K. This results in a theoretical change of the transition temperature of 0.44 K for $\epsilon_a = 0.022\%$, which is in good agreement with our experimental results (figure 3(b)).
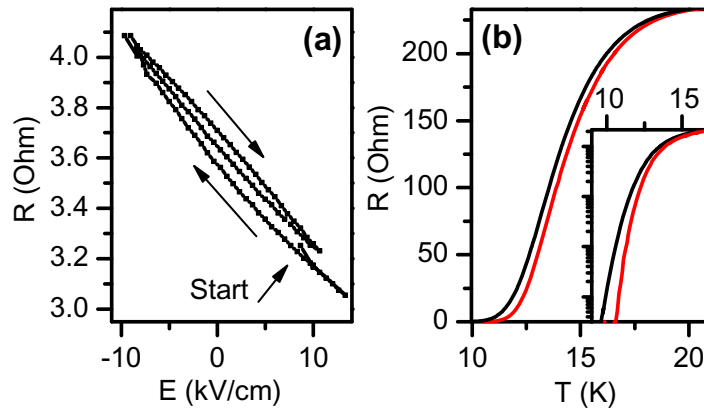
Figure 4. (a) Reversible change of resistivity over applied field at a fixed temperature of 18 K; (b) superconducting transition temperature of Ba-122 at $E = -6.6\,\text{kV cm}^{-1}$ (black) and $E = 6.6\,\text{kV cm}^{-1}$ (red). The inset shows the logarithmic scaled resistivity.

We checked the reversibility of the applied strain and the relaxation time of the PMN-PT at low temperatures, which is the time a piezoelectric material needs to reach the equilibrium strained state. We measured the resistivity, depending on the applied electric field at a fixed temperature within the superconducting transition at 18 K. At this point the slope is very steep, enabling the detection of minor changes in resistivity when the transition curve is shifted. Starting at $E = 8.66\,\text{kV cm}^{-1}$, we determined the resistivity by varying the field in steps of $0.66\,\text{kV cm}^{-1}$. By successive change of the electric field, a reversible change in resistivity was obtained (figure 4(a)). We attribute the minor deviation from linear behavior to the fact that the time between field change and data acquisition was too less to reach the equilibrium strain state. To characterize the relaxation time, we reversed the polarity of the electric field starting at $13.3\,\text{kV cm}^{-1}$ within 5 s and subsequently measured the resistivity, depending on time. Choosing a criterion for the equilibrium of less than 1% resistance change per hour, equlibrium is reached after 30 min.

Applying an electric field to an STO-buffered Ba-122 film, we observed a shift of the superconducting transition of 0.2 K for $\epsilon_a = 0.017\%$ (figure 4(b)) corresponding to $\beta = 1700\,\text{K}$. We compared the data with the results on statically epitaxial strained Ba-122 thin films where the compressive strain results in different c/a ratios. There a strain of $\epsilon_a = 1.2\%$ was achieved, resulting in a shift of the critical temperature of about 8 K [10]. The corresponding $\beta = 670\,\text{K}$ is less than half the value we achieve with the dynamic approach. Analyzing this difference, one has to take into account that equation (1) is only valid for small strain effects. In addition, hydrostatic pressure experiments reveal a nonlinear change in T_c with pressure [1]; we expect a similar behavior for biaxial strain.

In summary, we demonstrated the suitability of the inverse piezoelectric effect for the dynamical investigation of strain-dependent superconducting properties. We observed a significant change in the superconducting transition temperature for both LSCO and Ba-122 thin films. We conclude that compressive biaxial strain enhances the critical temperature for Ba-122 similar to cuprates such as LSCO.

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