Novel method to study strain effect of thin films using a piezoelectric-based device and a flexible metallic substrate

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To apply tensile or compressive uniaxial strain to functional thin films, we propose a novel approach in combining a piezoelectric-based device and a technical metallic substrate used widely in second generation coated conductors (i.e. superconducting tapes). A strain-induced shift of the superconducting transition temperature of 0.1 K for Co-doped BaFe2As2 was observed along the [100] direction, corresponding to a uniaxial pressure derivative \( dT_C/d\xi_{100} = -4 \) K/GPa. For Mn3CuN, a uniaxial strain derivative along the [100] direction of the Curie temperature \( dT_C/d\xi_{100} = 13 \) K/K was observed. The current approach is applicable to various functional thin films in a wide range of temperatures.

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Exploring the uniaxial pressure dependence of physical properties of functional materials gives an important clue for the fundamental understanding of physics. To conduct such experiments, single crystals with a reasonable volume are preferable. For some functional materials, however, the growth of sizable single crystals can be problematic, even though epitaxial thin films can be grown, e.g. Fe-based superconductors represented by NdFeAs(O,F), SmFeAs(O,F), and antiperovskite manganese nitrides, Mn3CuN. Moreover, non-equilibrium phases, e.g. (Ba,RE)Fe2As2 (RE: rare earth elements = La, Ce, Pr and Nd) and FeSe1-xTe, with low Te content are stabilized in the form of thin films. However, external pressure hardly transfers to strain in a thin film, since thin films are usually fabricated on substrates with high rigidity.

A common way to apply pressure to epitaxial films is to employ the lattice mismatch between the substrate and the film. But this method often (or strictly speaking always) requires the film thickness to be less than the critical value for which the coherent growth is maintained. Beyond the critical thickness, misfit dislocations or even cracks are introduced to partially relax the lattice stress, which may mask the interplay between the lattice strain and physical properties.

An alternative approach is to grow epitaxial thin films on piezoelectric substrates and to observe the changes in physical properties successively and reversibly by dynamic biaxial in-plane strain using the inverse piezoelectric effect. For instance, the Curie temperature of La1.85Sr0.15CuO4 and 0.2 K for BaFe1.8Co0.2As2 were observed to 0.022% at 20 K. Here, the in-plane strain \( \epsilon_{100} \) and \( \epsilon_{010} \) are defined by \( \epsilon_{100} = \epsilon_{\text{strain1}}/\epsilon_{0} \) and \( \epsilon_{010} = \epsilon_{\text{strain2}}/\epsilon_{0} \), respectively.

To overcome those problems and also to investigate the effect of in-plane symmetry breaking on the physical properties, we report on a novel approach in combining a piezoelectric-based device and a technical metallic substrate used widely in second generation coated conductors (i.e. superconducting tapes). The device has one central and two outer piezostacks as shown in Fig. 1(a). The central stack and the outer stacks generate compressive and tensile strains or vice-versa [Fig. 1(b)]. Since the piezostacks are longer than the effective sample length (see below), large stress can be applied to the sample even at low temperatures. Additionally, the technical substrate is flexible thanks to its low thickness of 100 µm, resulting in a high ability to transfer large amounts of strain to the film. Besides, MgO templates prepared on the technical substrate by ion beam assisted deposition (IBAD) are suitable for epitaxial growth of various materials. Hence, our approach is applicable to a wide range of functional materials, whenever epitaxial thin films are realized on IBAD-MgO Hastelloy. Indeed, Ref. 12 have fabricated Nd2Fe14B thin films on Mo-buffered IBAD-MgO Hastelloys and strained the substrate plastically (\( \epsilon \approx 2\% \)) by an external load which significantly influenced the magnetic properties of the film. Our approach extends this concept and allows dynamic and reversible control of the in-plane uniaxial strain. In this study, an Fe-based superconductor, Co-doped BaFe2As2, and a magnetostRICTive material, Mn3CuN, have been chosen as test materials.

The uniaxial strain apparatus [Fig. 1(a)] composed of three piezostacks [Pb(Zr,Ti)O3], Piezomechanik GmbH] was constructed according to Ref. 11. A strain gauge was mounted on the backside of the device. The commercially available technical metallic substrates on which biaxially textured MgO templates of 160 nm thickness were deposited by IBAD were provided by iBeam Materials, Inc. To avoid inter-diffusion between the metallic tape (Hastelloy C-276) and MgO, the template contains a Y2O3 layer of 1 µm thickness. The thermal expansion coefficient of Hastelloy, Y2O3, and MgO are 1.08 × 10^{-6}/K, 1.13 × 10^{-6}/K, and 1.24 × 10^{-6}/K, respectively, indicating that the thermal expansion mismatches between Hastelloy and Y2O3 and between Y2O3 and MgO are not trivial. However, the Y2O3 layer is polycrystalline and has a large thickness of 1 µm. Therefore, the strain due to thermal expansion mismatch is negligible.

The technical substrate was cut into a small slab of 3.5 mm length and 1 mm width by laser cutting, which was used as a test piece. The test piece was bridged across the gap between...
the two sample plates using Stycast 2850FT, as shown in Fig. 1(a). The gap between the sample plates, i.e. the effective length of the test piece, was around $L = 2.0 \text{ mm}$. By applying a bias voltage of $\pm 30 \text{ V}$ at room temperature, displacements of around $1.7 \mu \text{m}$ and $-2.0 \mu \text{m}$ were observed, respectively [Fig. 1(c)], which corresponds to a maximum change in strain of $0.18\%$. As expected, the displacement was significantly reduced to $\pm 0.7 \mu \text{m}$ at $T = 14 \text{ K}$ even when applying a bias voltage of $\pm 110 \text{ V}$, corresponding to a strain level of $1.4/2000 \times 100 = 0.07\%$. However, this level of strain is large enough for observing shifts of $T_c$ for Co-doped BaFe$_2$As$_2$.

Co-doped BaFe$_2$As$_2$ thin films were deposited on IBAD-MgO Hastelloy at $800 \degree \text{C}$ by pulsed laser deposition (PLD) using a Nd:YAG 3rd harmonic laser ($\lambda = 355 \text{ nm}$) under ultra-high vacuum condition. This might be the first report ever on using a third harmonic Nd:YAG for depositing BaFe$_2$As$_2$. The nominal composition of the sintered pellet used as a PLD target was Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$. The energy density was around $4 \text{ J cm}^{-2}$ at the surface of the target. Figure 2(a) shows the $\theta/2\theta$-scans of the Co-doped BaFe$_2$As$_2$ (Ba-122) film on IBAD-MgO Hastelloy. All peaks except for a small amount of an unknown phase can be indexed with Co-doped Ba-122, MgO, Y$_2$O$_3$ and Hastelloy. The reflections from Co-doped Ba-122 were only 00$l$, indicative of the $c$-axis being oriented normal to the technical substrate. The azimuthal $\phi$-scan of the off-axis 103 reflection of Co-doped Ba-122 showed four peaks [Fig. 2(b)]. These results prove that Co-doped BaFe$_2$As$_2$ was grown on IBAD-MgO Hastelloy with cube-on-cube configuration. The FWHM value ($\Delta \phi$) of the 103 reflection was around 4°, which is similar to the underlying MgO. The films were cut into rectangular slices (5 mm long and 1 mm wide) by laser cutting. The longer direction is defined as being parallel to the crystallographic [100] direction.

Mn$_3$CuN thin films with 80 nm thickness have been fabricated on IBAD-MgO templates at 200 °C in a custom-designed reactive sputtering chamber equipped with a high magnetic field of $\sim 1 \text{ T}$ at the surface of the target. Mn/Cu = 4:1 alloy was employed as the target, and sputtereng was conducted in an Ar/N$_2$ gas mixture. The as-grown film was sealed together with Ti powder as an oxygen absorber in a quartz tube, which was evacuated to a pressure level of $1 \times 10^{-4} \text{ Pa}$. The whole arrangement was heated to 650 °C at a rate of 200 °C min$^{-1}$ and subsequently furnace-cooled to room temperature. The process details can be found in Ref. 4. Structural characterization for the resultant film by X-ray diffraction is summarized in Fig. 3. In Fig. 3(a), the 002 and 004 reflections of Mn$_3$CuN together with the 200 reflection of MnO were detected besides the peaks related to the technical substrate (i.e. Y$_2$O$_3$, MgO and Hastelloy). Although Ti powder was very effective at avoiding oxide formation in the post-annealing process, a small amount of MnO has formed. It is plausible that the oxygen came from the Y$_2$O$_3$ buffer layer. Nevertheless, the presence of MnO in our film does not affect the current investigation. Shown in Fig. 3(b) is the $\phi$-scan of the 111 reflection for Mn$_3$CuN. A clear four-fold symmetry was observed. These results prove that Mn$_3$CuN can be grown epitaxially on IBAD-MgO Hastelloy. After structural characterization, the films were cut into rectangular slices in the same way as the Co-doped BaFe$_2$As$_2$ thin films.
Figure 4 shows the shift of the resistance curves for Ba(Fe0.92Co0.08)2As2 thin films on IBAD-MgO Hastelloy by uniaxial strain along the [100] direction. The offset $T_c$ defined as the intersection between the steepest slope of the resistivity curve and zero resistance were 14.2 K for tensile ($\varepsilon_{100}=0.018\%$) and 14.3 K for compressive strain ($\varepsilon_{100}=-0.03\%$), respectively. This tiny change is due to the small sensitivity of $T_c$ against the strain along the [100] direction. The strain-direction dependence of $T_c$ for P-doped Ba-122 single crystals showed that the change in $T_c$ is smallest for the [100] direction compared to the [110] and [001] directions.\(^{20}\) A similar trend is expected for Co-doped Ba-122, although the doping element differs from P. On the assumption that the Young modulus ($E_{100}$) of Co-doped Ba-122 is 55 GPa,\(^{21}\) the respective tensile and compressive stress values are calculated as $9.9 \times 10^{-3}$ GPa and $-1.65 \times 10^{-2}$ GPa using Hooke’s law. Accordingly, the uniaxial pressure derivative of the offset $T_c$ approximates $dT_c/d\varepsilon_{100}$ along [100] without considering orthorhombic distortion is calculated as 13 K/%.

It is reported that the $T_c$ of epitaxial Mn3CuN thin films grown on different substrates systematically changes due to the biaxial strain.\(^{4}\) The strain dependence of $T_c$ without considering a Jahn–Teller strain is described by

$$T_c = T_c(0) + (2\varepsilon_{100} + \varepsilon_{001}) \frac{dT_c}{d\varepsilon_{100}}$$

where $T_c(0) = 143$ K is the Curie temperature of unstrained Mn3CuN.\(^{25}\) Using the reported values of $T_c$ and the lattice parameters at room temperature for films on different substrates,\(^{4}\) the strain derivative is evaluated as $dT_c/d\varepsilon_{100} = 48$ K/%. It is expected that this value does not change significantly near the transition temperature, since the linear thermal expansion coefficient of Mn3CuN ($1.77 \times 10^{-5}$/K)\(^{23}\) is comparable to that of MgO ($1.24 \times 10^{-5}$/K).\(^{17}\) Nevertheless, the strain derivative derived from the study

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**Fig. 3.** (Color online) Summary of the structural characterization for Mn3CuN fabricated on the IBAD-MgO template. (a) The $\theta$2$\theta$-scan and (b) the azimuthal $\phi$-scan of the off-axis 111 reflection of Mn3CuN.

**Fig. 4.** (Color online) (a) The resistance curves for BaFe$_{0.92}$Co$_{0.08}$As$_2$ thin films under various uniaxial strain state. (b) Enlarged view of the resistance curves in the vicinity of zero resistance.
approach thus greatly expands the experimental range of many functional thin films in allowing investigations on strain-driven changes in physical properties.

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Fig. 5. (Color online) (a) The normalized resistivity curve for the relaxed Mn$_3$CuN thin film. (b) The resistivity curves for the Mn$_3$CuN thin film under various uniaxial strain states. The inset shows an enlarged view of the resistivity curves in the temperature range 150 ≤ T ≤ 170 K.

using single crystalline substrates is notably different from the results obtained from the uniaxial strain in this study. The reason for this discrepancy is not clear but might be due to nitrogen deficiency and/or off-stoichiometry. Further studies are necessary to elucidate the cause of this discrepancy.

In summary, we have proposed a novel approach combining a piezoelectric-based apparatus and flexible metallic substrates for investigating the in-plane uniaxial pressure dependence of the physical properties of functional thin films. The strain-induced shift of $T_c$ along the [100] direction was around 0.1 K for Co-doped BaFe$_2$As$_2$, corresponding to the uniaxial pressure derivative $dT_c/dp_{100} = -4$ K/GPa. For the Mn$_3$CuN film, the uniaxial strain derivative $dT_c/d\varepsilon_{100}$ of the Curie temperature along the [100] direction was 13 K/%. Our novel method demonstrates that uniaxial strain dependence of the physical properties of functional thin films can be investigated dynamically and reversibly, whenever epitaxial thin films are realized on IBAD-MgO Hastelloy.

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