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Structural properties of superconducting $Ba(Fe,Co)_2As_2$ thin films

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Introduction

- Iron-based superconductors exhibit large, nearly isotropic critical fields at low temperatures [1]
- Superconducting thin films [2] are of interest for their fundamental properties and applications like superconducting tapes [3]
- Superconducting properties like transition temperature T_c, upper critical field strength H_{c2} and critical current density J_c depend strongly on microstructure
- Co-doped $BaFe_2As_2$ (Ba122) is a widely studied model system (Fig. 1)
- Growth mechanism of Ba122 on single crystalline substrates is not yet fully understood due to different fabrication parameters and possible interactions with the substrate [4]

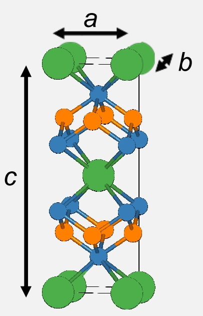

Fig. 1: Tetragonal unit cell of $BaFe_2As_2$ with $a = b = 3.96$ Å and $c = 13.02$ Å.

Goals

- Microstructural and chemical analysis of Ba122 in dependence of fabrication parameters and substrate material
- Understanding possible interaction of Ba122 with the substrate and the electron beam (i.e. beam damage)
- Correlation of microstructure and superconducting properties

Sample fabrication

- $Ba(Fe_{1-x}Co_x)_2As_2$ thin films with nominal doping of $x = 0.08$ and varying thickness (20 – 100 nm) were deposited on heated (700 – 750 °C) single-crystalline CaF_2, $LaAlO_3$ and MgO substrates (room temperature in-plane lattice mismatch of 2.6% (compressive), 4.6% (compressive) and 5.9% (tensile), respectively) by pulsed laser deposition
- Cross-section samples were prepared using the focused Ga-ion beam *in-situ* lift-out technique (final polishing at 5 keV) with an FEI Strata 400S dual-beam system

Experimental techniques

- An image C_s-corrected FEI Titan³ operated at 300 kV was used to analyze the microstructure with high-resolution (scanning) transmission electron microscopy (HR-(S)TEM) and to apply core-loss electron energy loss spectroscopy (EELS) with a Gatan Tridiem 865 imaging filter
- (High-angle) annular dark-field ((HA)ADF) STEM was used for material (*Z*-) contrast imaging
- Energy-dispersive X-ray spectroscopy (EDS) was performed on an FEI Tecnai Osiris operated at 200 kV and equipped with ChemiSTEM technology
- The signal-to-noise ratio of EELS and EDS spectrum images was improved by principal component analysis (HyperSpy / temDM MSA [5]) before extracting qualitative elemental maps (Cornell Spectrum Imager [6]/Digital Micrograph)
- Spectroscopic measurements of Ba122 on CaF_2 were mainly performed by EELS to avoid overlap of F K_α (677 eV) and Fe L_α (705 eV) in EDS

Results

- Ba122 shows layered, epitaxial growth on CaF_2 and $LaAlO_3$ and polycrystalline structure on MgO (cf. Fig. 2a-c)
- An amorphous O-rich layer is observed on top of the Ba122 layer (marked in Fig. 2a)
- Stacking faults (SFs) on the Ba-planes show dark contrast in HAADF-STEM images
  → Most SFs present in Ba122 on $LaAlO_3$ (Fig. 2b and 3)
  → Dark contrast (Fig. 2f) may arise due to dechanneling of electrons around defective region, presence of vacancies and/or outdiffusion of Ba and indiffusion of lighter elements (e.g. O)
- EELS reveals presence of O at SFs (Fig. 3)
- Ba122-CaF_2-interface is susceptible to beam damage in conventional TEM (Fig. 2d)
  → Formation of an amorphous layer („damaged region“)
  → STEM imaging gives better control over distributed dose and reduces beam damage
- Formation of BaF_2 at the Ba122-CaF_2-interface is visible in HR-STEM images (Fig. 2e) as determined by Fourier-transform analysis of the crystal structure
- Fe-rich precipitates are observed in all samples (cf. Fig. 4b,c, not shown for CaF_2)
- Additionally, O-rich phases showing dark HAADF-STEM contrast are observed in Ba122 on CaF_2 (Fig. 4a) which are currently under investigation

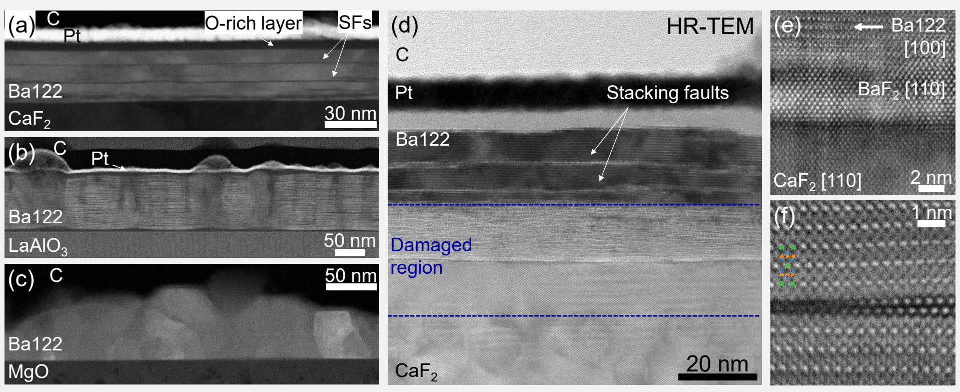

Fig. 2: HAADF-STEM overview images of Ba122 layers on (a) CaF_2, (b) $LaAlO_3$ and (c) MgO. (d) HR-TEM image showing formation of an amorphous layer between CaF_2 and Ba122 under the electron beam. HAADF-STEM images of (e) the Ba122-CaF_2-interface and (f) a SF.

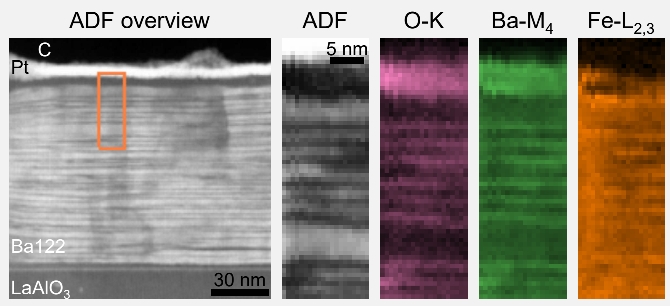

Fig. 3: ADF image and mappings of EELS core-loss signals for Ba122 on $LaAlO_3$ from the marked region in the overview image. The SFs show dark contrast in the ADF image, an increased O and Ba signal and a reduced Fe signal.

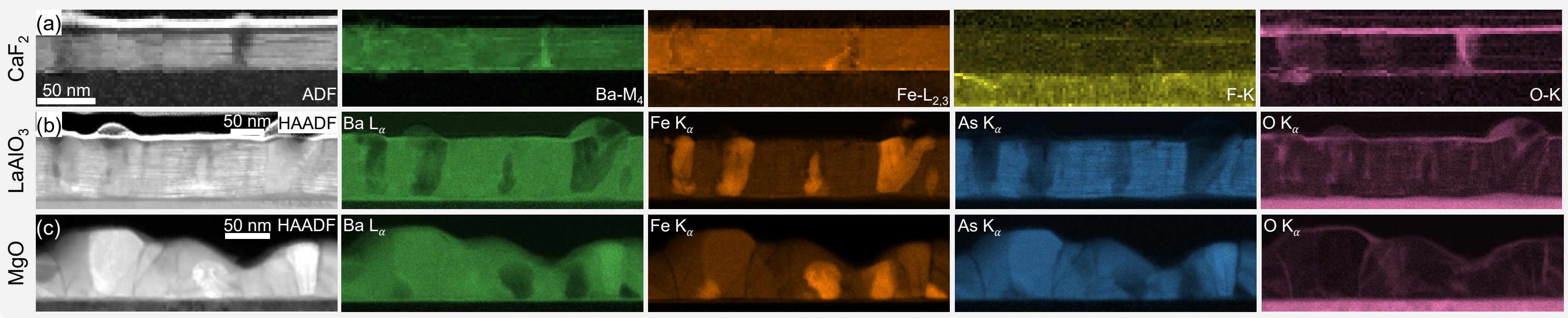

Fig. 4: (a) Mappings of EELS core-loss signals for Ba122 on CaF_2. EDS signals for Ba122 on (b) $LaAlO_3$ and (c) MgO show the presence of Fe-rich precipitates. An enhanced O signal is found at the film surface, near precipitates and grain boundaries.

Summary

- The investigated Ba122 layers contain (Fe-rich) precipitates and stacking faults, which are less frequent for layers with smaller lattice mismatch to the substrate
- The cleanest Ba122 phase is found on CaF_2 which is consistent with highest measured T_c of 23 K among the shown samples
- High electron doses can lead to destruction of Ba122 layer, especially on CaF_2

References

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