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Pulsed Laser Deposition of quasi-multilayer superconducting Ba(Fe_{0.92}Co_{0.08})₂As₂-BaHfO₃ nanocomposite films

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Abstract. Quasi-multilayer films with perovskite nano-particles have already been investigated and reported in YBCO compounds. Here, we introduce this technique to iron based superconductors by preparing quasi-multilayer nanocomposite films of Co-doped BaFe₂As₂ with BaZrO₃ and BaHfO₃ perovskite nanoparticles by a pulsed laser deposition technique using a frequency-tripled Nd:YAG laser ($\lambda = 355$ nm) with separate targets of the constituents. We investigated the enhancement of pinning force density and critical current density in relation to critical temperature reduction. Application-relevant properties are discussed in relation to the deposition conditions and the microstructural properties of the films to better understand growth and behavior of strong pinning centers in these materials.

1. Introduction

An interesting development in solid state physics was the discovery of the iron-based superconductors (FBS) [1] with transition temperatures up to 55K in 2008. In this class of superconductors, the FeAs or FeSe [2, 3] layers carry the superconductivity comparable to CuO planes in cuprates. With their properties they can be seen as in between cuprates (layer-type crystal structure, high upper critical fields, 2D-behaviour), the low temperature superconductors, such as NbTi and Nb₃Sn [4] (few thermal fluctuations, low anisotropies) and magnesium diboride (multi-band superconductivity). This combination of different properties, especially low anisotropies and high critical fields, makes iron-based superconductors an interesting candidate for high field applications. For that, the production of long tape samples must be reliable and affordable, flux pinning mechanisms must be understood correctly and the transport properties must be adjustable to all needs of applications. The research is still at the beginning and requires several model experiments on thin films. The realization and characterization of nano-structured thin films for pinning is a good example because of multi-phased, self-organizing heteroepitaxial layer growth.

Quasi-multilayered films made via pulsed laser deposition (PLD) with two targets by implementing nano-particles of BaHfO₃ (BHO) [5, 6] and BaZrO₃ (BZO) [7, 8] have been already reported for YBCO-compounds. Films made with mixed Co-doped Ba122 and BZO targets were also already studied [9] and published in 2017 as well as films with mixed P-doped Ba122 and BZO by Miura et al [10, 11].



In this study, we combine the multi-target technique for the fabrication of FBS thin films since this has the advantage of easy adjustment and control of the doping level during deposition with the usage of BHO as nano-particle material.

2. Experimental

We prepared nanocomposite films of $\text{Ba}(\text{Fe}_{0.92}\text{Co}_{0.08})_2\text{As}_2$ films with BHO nanoparticles with film thicknesses between 35 and 110 nm by a quasimultilayer pulsed laser deposition technique using a Quanta-Ray INDI-40 frequency-tripled neodymium-doped yttrium aluminum garnet (Nd:YAG) laser ($\lambda=355$ nm) by Newport Spectra-Physics GmbH with separate targets of the constituents with a maximum diameter of 1 inch. Films were deposited on CaF_2 (100) single crystalline substrates in ultra-high vacuum conditions ($p = 10^{-9}$ mbar) with a pulse duration below 10 ns and a maximum repetition rate of 10 Hz in the temperature range of 700-750 °C. We used laser energy of 3.0 J/cm². The substrate-target distance was 40 mm. Deposition parameters are computer-controlled to obtain best reproducibility. The target diameter of the $\text{Ba}(\text{Fe}_{0.92}\text{Co}_{0.08})_2\text{As}_2$ is 10 mm and 1 inch for the BHO. The $\text{Ba}(\text{Fe}_{0.92}\text{Co}_{0.08})_2\text{As}_2$ targets were prepared according to ref. [12] at IFW Dresden. The BHO targets were prepared at Institute for Technical Physics of the Karlsruhe Institute of Technology from BaCO_3 and HfO_2 powder. The powders are mixed, pressed and sintered up to 1150 °C. The result is ground and pressed again in an isostatic press and sintered again up to 950 °C. The film thickness was determined by atomic force microscope (AFM) measurements. The crystalline structure of the films was investigated via x-ray diffraction (XRD) (Bruker D8, $\text{Cu K}\alpha$ $\lambda = 1.54$ Å). Application-relevant properties, such as critical current density J_c and according pinning force density F_p as well as the critical temperature T_c were determined from temperature-dependent electrical measurements using a 14 T Physical Property Measurement System (PPMS) by Quantum Design. The chemical composition of the films was investigated with Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Additional (scanning) transmission electron microscopy ((S)TEM) was used to determine the film thickness, size and shape and the distribution of the nano-particles after the deposition and to calibrate the parameters for further depositions in the future.

3. Results and discussion

Previous to the quasi-multilayered films we optimized the deposition parameters of $\text{Ba}(\text{Fe}_{0.92}\text{Co}_{0.08})_2\text{As}_2$ on CaF_2 with our deposition setup regarding T_{c0} and J_c to ensure high reproducibility of the superconducting films. We used single-crystalline CaF_2 (100) with a size of 10 by 10 mm. The best films showed a T_{c0} of 24 K [13]. We began using the quasi-multilayer technique by implementing 2 mol% of BZO into the matrix and were able to show that nano-particles (see Figure 1) are forming similar to the films from Lee et al. [9].

After that finding several quasi-multilayered films with different doping levels of BHO (0 mol%, 0.18 mol%, 0.69 mol% and 1 mol%) were prepared and investigated. The total number of $\text{Ba}(\text{Fe}_{0.92}\text{Co}_{0.08})_2\text{As}_2$ pulses of 18000 was not changed in order to keep the (superconducting) film thickness constant. The total amount of BHO was controlled by the number of BHO interlayers of constant 12 pulses BHO per layer. For 0.18 mol% we exchanged targets 9 times and added a cap layer of 1800 pulses of Ba122. The number of exchanges for 0.69 mol% is 39 and the Ba122 cap layer consists of 450 pulses. For 1 mol% of BHO we had 143 exchanges and a cap layer of 125 pulses. The layer thickness of the films was in the range of 35 to 110 nm. As shown in Figure 2 at a doping level of 2 mol% of BHO we were able to show that nano-particles form in the Ba122 matrix just as for BZO.

The θ - 2θ scans in Figure 3 indicate c -axis texture of each film. The pristine film shows some secondary phases at 33° and 67°. The original structure of the $\text{Ba}(\text{Fe}_{0.92}\text{Co}_{0.08})_2\text{As}_2$ is throughout the different doping levels still preserved. A slight hint of a BHO peak can be observed for 1% BHO at an angle of around 30°. The peaks tend to shift around 0.5 ° at a doping level of 1 mol% to higher angles which indicates a reduction of the c -axis parameter (1.31097 nm to 1.31972 nm) caused by strain introduced by the BHO particles in the film.

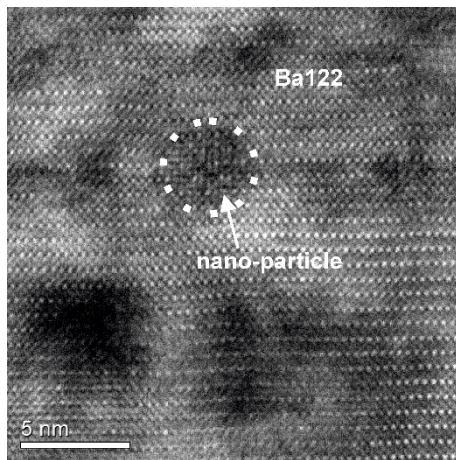


Figure 1. HR-STEM image of a sample with 2 mol% of BZO in the Co-doped Ba122 matrix

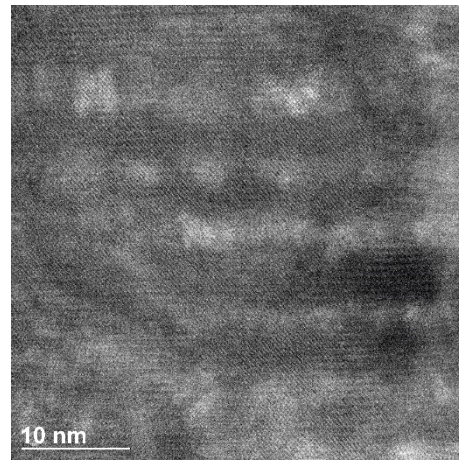


Figure 2. HR-STEM image of a sample with 2 mol% of BHO in the Co-doped Ba122 matrix

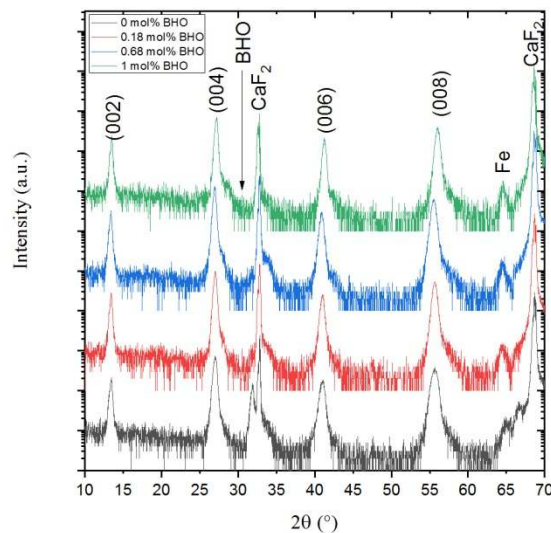


Figure 3. θ - 2θ scans of $\text{Ba}(\text{Fe}_{0.92}\text{Co}_{0.08})_2\text{As}_2$ films grown with different doping levels.

The T_{c0} values (see Figure 4) are decreasing with increase of the doping level as expected and reported before [14]. Just for a very low doping level is T_{c0} slightly above the value for the pristine $\text{Ba}(\text{Fe}_{0.92}\text{Co}_{0.08})_2\text{As}_2$ film. The difference is only 0.9 K and within our usual experimental scatter.

The critical current density J_c reveals the expected behavior of the superconducting films caused by doping. The critical current density J_c increases significantly with increasing doping level and reaches almost 10^6 A/cm² for the highest doping level of 1 mol% and an external magnetic field of 0 T. Due to low growth rates, J_c and T_{c0} of the pristine film is relatively low but are the optimum which can be achieved with our deposition setup.

The pinning force F_p also increases with the doping level and reaches 50.6 GN/m³ at 11T and a doping level of 1 mol%. Considering the relatively low BHO concentration in our study, this value agrees well literature data on BaZrO₃-added $\text{Ba}(\text{Fe},\text{Co})_2\text{As}_2$ (170 GN/m³ for 2mol% and 30 GN/m³ for 4mol% at 11 T, 4.2 K [9] and $\text{BaFe}_2(\text{As},\text{P})_2$ films (58 GN/m³ at 9 T, 5 K [10] and recently 189 GN/m³ at 9 T, 4 K [11]).

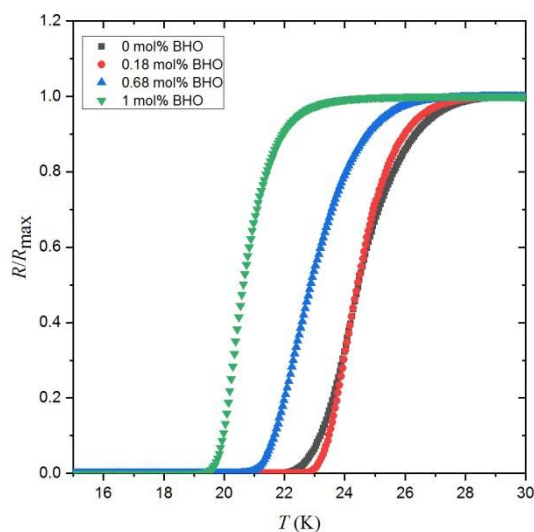


Figure 4. Temperature dependence of the resistance normalized to the value above T_c .

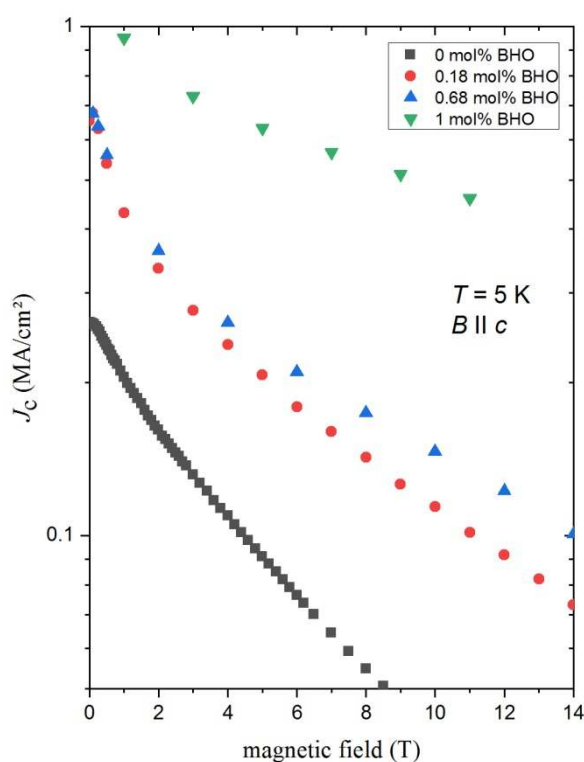


Figure 5. Critical current density J_c plotted as a function of the external magnetic field.

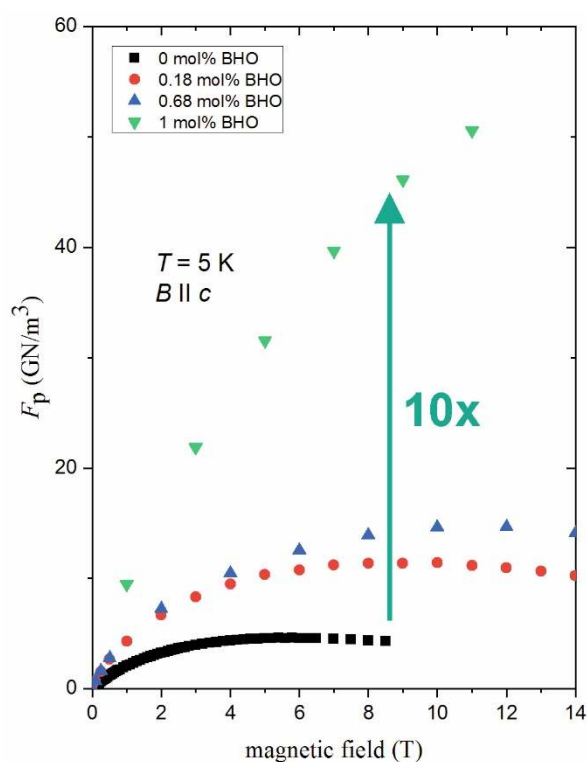


Figure 6. The pinning force density F_p plotted as a function of the external magnetic field.

4. Conclusion and outlook

Finally, we showed the preparation of the first quasi-multilayered iron-based superconducting films with perovskite nanoparticles as well as J_c improvement with BHO nanoparticles in BaFe₂As₂ films. The results show a similar behavior as the results for YBCO or for iron-based superconducting films with

premixed dopants. We found promising results to probably introduce to applications in the future such as tapes and powder-in-tube (PIT) cables [15].

Next step in investigating pinning properties is to perform several investigations to achieve a proper calibration of our parameters to be able to adjust the doping level before the deposition to manufacture superconductors with designed properties. For that we have to change the film and layer thicknesses as well as the frequency of target exchange during deposition. The size and shape of the nano-particles have to be investigated intensively. The strong T_c reduction will be part of further investigations as well.

Further experiments will also be performed with the usage of BZO according to the results presented in this work.

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