

Preparation and Investigation of High-Temperature Superconducting Thin Films In the Superconductivity and Cryoelectronics Laboratory of the Institute of Electronics Bulgarian Academy of Sciences

T. Nurgaliev

Institute of Electronics BAS, 72 Tsarigradsko Shose Blvd, 1784 Sofia, Bulgaria
timur@ie.bas.bg; www.ie-bas.dir.bg



Abstract. Thin films of two high-temperature superconducting materials ($\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{C}_3\text{O}_x$) were prepared by radio frequency magnetron sputtering in Institute of Electronics, Bulgarian Academy of Sciences in 1988. *Superconductivity and cryoelectronics* laboratory was created (1989) for the further developing of the superconductivity topic in the Institute of Electronics. This paper describes some stages of the development of the investigations of high-temperature superconducting thin films and layered structures in the *Superconductivity and cryoelectronics* laboratory.

Keywords: HTS thin films, YBCO, BSCCO, Magnetron sputtering

1. Introduction

K. A. Müller and J. G. Bednorz discovered in April 1986 that a ceramic compound $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ became superconducting at 35 K. In 1987 two groups independently reported critical temperature well above the temperature of liquid nitrogen (77°K) in the ceramic compound $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO). These ceramics were specified as high-temperature superconductors (HTS). Research on high-temperature superconductivity materials started in Bulgaria almost immediately after discovering the phenomenon. Bulk high-temperature superconductivity ceramic materials were prepared and Institute of Solid State Physics BAS, Sofia University “St. Kliment Ohridski”, and Institute of Electronics – Bulgarian Academy of Sciences etc started to investigate their properties [1-5].

2. Institute of Electronics BAS

The Institute of Electronics BAS possesses potential and qualified specialists in the field of superconductivity (such as young PhD S. Tinchev, Z. Ivanov, B. Todorov, which received their degrees in Moscow State University, Russia) to develop the high-temperature superconductivity topic. Institute of Electronics BAS started to develop Bulk high-temperature superconducting technologies (I. Nedkov). By taking into account the importance of thin films of high-temperature superconducting materials for microelectronics applications, the experiments on magnetron sputtering (B. Goranchev, Z. Ivanov, B. Todorov, T. Donchev), electron-beam evaporation (G. Mladenov) and laser ablation (P. Atanasov) of YBCO materials were started as well. Thin films of high-temperature superconductivity $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{C}_3\text{O}_x$ materials were prepared by radio frequency magnetron sputtering in Institute of Electronics BAS (1988) [6-7].

3. Superconductivity and Cryoelectronics Laboratory and Development of Thin Film Technologies

For the further developing of the superconductivity topic, the Scientific Council of the Institute of Electronics BAS made decisions (from 26.10.1989 and 16.11.1989) to form a special laboratory in the institute. Therefore, 27.11.1989 the *Superconductivity and*

cryoelectronics laboratory (with Head – Professor A. Spasov) has formed and seven scientists joined this laboratory.

High-temperature superconducting thin films: For developing of high-temperature superconducting thin film deposition technologies two Universal Vacuum Posts (VUP 5) and a radio frequency magnetron with its feeding source were bought for the new laboratory in 90th of the 20 century. A new configuration of a two opposing – the targets direct current magnetron sputtering system (T. Donchev) has built (Fig. 1) as well. The new equipment and the usage of good quality YBCO targets allowed to successfully develop the technologies for in-situ deposition of epitaxial thin high-temperature superconductivity $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) films on MgO , LaAlO_3 (LAO), SrTiO_3 (STO), Y- ZrO_2 (YSZ) and Si substrates with and without buffer layers (V. Tsaneva, T. Donchev, R. Chakalov). The qualities of these films grown on MgO , LAO and STO substrates were very high and the critical temperature T_C and the critical current density J_C were not less than 90°K and 10^6 A/cm^2 (at 77°K), respectively [8-9].

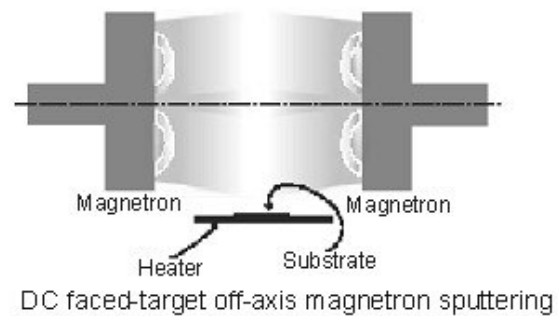
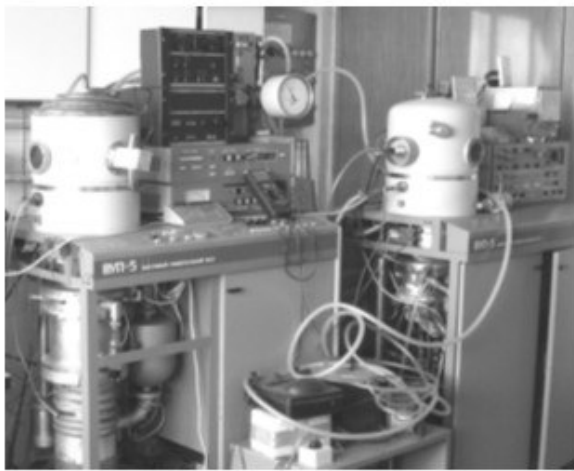


Fig.1 Radio frequency (left) and two opposing – targets DC magnetron sputtering (right) systems for obtaining HTS and ferromagnetic manganite films and dielectric buffer layers.

Optical emission spectroscopy was employed as an additional non-perturbing method for investigation and controlling of the condensing species flux during high-temperature superconductivity and buffer thin film deposition process (V. Tsaneva) [10]. The oxygen-to-argon emission peaks ratio was shown to be a useful indicator for controlling of the oxygen partial pressure and the substrate temperature in order to obtain smooth epitaxial films with high critical parameters.

Later direct current magnetron sputtering was used to prepare YBCO films on both sides of the substrates (double-side films) [11]. Nearly identical values of the characteristics of two-sided films were obtained by a careful optimization of the deposition condition.

A high-temperature superconducting YBCO films on miscut NdGaO_3 substrates (or tilted substrates) [12] were prepared as well (Fig. 2). Films grown on the substrates with the optimal tilt angle demonstrated greater values of the critical temperature and the critical current density and smaller value of the microwave surface resistance.

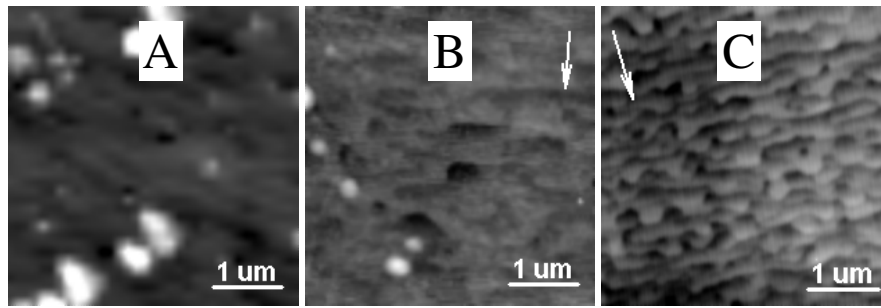


Fig. 2 AFM image of the surface of YBCO thin films deposited on NdGaO_3 substrates with the tilt angles 0° (A), 3° (B) u 5° (C)

Superconducting YBCO thin films prepared in the laboratory were used in numerous experiments in the Institute of Electronics at the Bulgarian Academy of Sciences as well as in the laboratories of the international scientific partners.

Ferromagnetic manganite films. In the 90s of the 20 century the ferromagnetic manganites (for example $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ (LCMO) and $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ (LSMO)) attracted the attention of researchers due to the colossal magnetoresistance effect. Thin films of LCMO were deposited by radio frequency magnetron sputtering under different conditions and their magneto-resistive properties were investigated in collaboration with the Institute of Solid State Physics BAS [13-14]. In collaboration with the same Institute technologies for radio frequency magnetron sputtering of thin films of more exotic materials as the orthorhombic multiferroic GdMnO_3 , semiconducting magnetic cobaltite $\text{NdBa}_2\text{Co}_2\text{O}_{5+x}$ were developed [15].

High-temperature superconductor – ferromagnetic manganite layered structures. High-temperature superconducting cuprate – ferromagnetic manganite (HTS/FM) structures are under intense study due to both a fundamental interest and the possible applications in spintronics, where the spin state of the charge carriers is used to control the device parameters. Therefore HTS/FM and FM/HTS double layer structures, consisting of YBCO and manganite layers were fabricated and their electrical parameters were investigated [16-17]. It was shown that i) the temperature dependence of the normal state resistivity of these HTS/FM structures is not described by a linear function; ii) the presence of a manganite layer leads to a worsening of the critical parameters of the YBCO film; iii) the manganite layer leads to a significant increase of the microwave surface resistance of the structure.

4. HTS Technology in Neighboring Laboratories

Laboratory Gas lasers and laser technologies – P. Atanasov: HTS YBCO thin films were prepared by in-situ oxygen plasma-assisted laser deposition. Low temperature (450 – 550 C) in-situ laser deposition in combination with CO_2 laser heating of the substrate and subsequent annealing in oxygen were shown to be a technique suitable for obtaining of oriented polycrystalline YBCO films with good superconducting properties. A multistep superfast CO_2 laser annealing in oxygen was proposed for modification of laser deposited YBCO films on Si. It was also shown that N_2 laser can be used for patterning YBCO thin films [18-19].

Laboratory Physical problems of electron beam technology – G. Mladenov:

Deposition of YBCO films using electron beam evaporation and simulation of the physical characteristics of the process were produced [20-21].

Laboratory Microwave magnetics – I. Nedkov: Investigation of the alkali metals (Na, K and Rb) influence on the properties of bulk high-temperature superconducting YBCO materials was performed [22-24]. Each impurity has a specific effect on the temperature dependence of the resistance and magnetic susceptibility.

5. Investigation of Characteristics and Application Possibilities of HTS Thin Films and Structures in the Superconductivity and Cryoelectronics Laboratory

Investigation of penetration of HTS films by inhomogeneous magnetic field and application for contactless characterization of HTS materials. An investigation was carried out of the interaction of spatially uniform direct current and alternating current magnetic fields and magnetic fields of cylindrical symmetries (magnetic field of small nearly planar coil with the current) with a thin superconducting film which has become partly resistive after the induced current density had exceeded the critical one (T. Nurgaliev) [8, 25]. A *Critical state model* was proposed for describing the penetration of the high-temperature superconducting film by non-uniform magnetic field of cylindrical symmetry [26]. The film remains in Meissner state, when the amplitude of the driving current is small in nearly planar coil. With an increase of the current amplitude in the coil the film is penetrated by the magnetic field and the trapped current appears near the mean radius of the coil windings. In this case the shielding current profile in the film depends not only on the drive current, but on the magnetic history of the film as well (Fig. 3) [28-30].

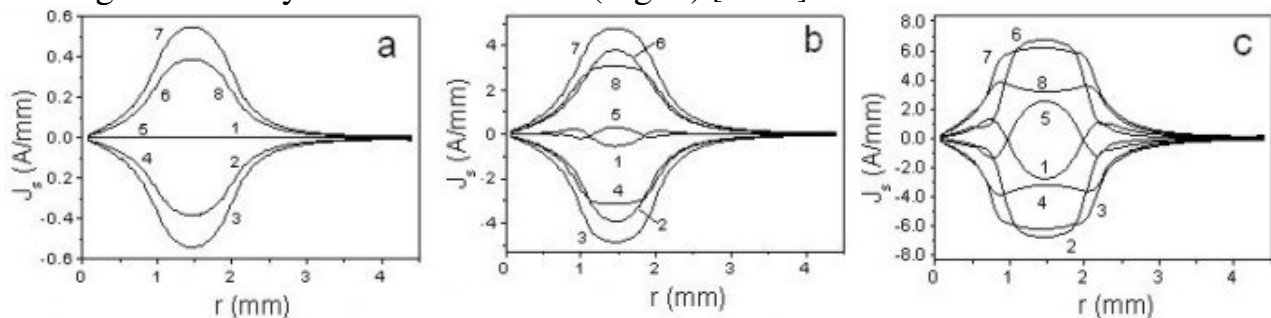


Fig. 3 Radial distribution of the screening current in HTS film, created by the magnetic field of a planar coil. The current amplitude in the coil is 0.01 A, 0.09 A and 0.15 A (a, b and c), and the phase of the current is $\tau = \pi, 1.25\pi, 1.5\pi, 1.75\pi, 2\pi, 2.25\pi, 2.5\pi, 2.75\pi$ (curves 1–8) [27].

The above results were used for the elaboration of contactless measuring set-ups of the parameters (voltage *versus* current curves, resistivity and critical current density) of thin high-temperature superconductivity films and of bulk high-temperature superconducting pellets [8, 23, 25-28]. This effectively contributed to developing of technology for growing of high quality high-temperature superconducting films in the laboratory.

Developing of investigations on Josephson junction in HTS materials and their device applications: One of the scientists in the laboratory (S. Tinchev) on a long-term leave at Forschungsgesellschaft fuer Informationstechnik (Germany) developed a technology for fabrication of 77 – K SQUIDS by local oxygen-ion bombarding. A non-uniform in-depth

distribution of the critical temperature was observed in the ion modified high- T_c YBCO film weak links. It was found that the film inside became granular after the modification. These SQUIDs were the first high $-T_c$ devices tested on a space mission on board of the space shuttle Discovery [29-30].

Z. Ivanov in his long-term stay in the Chalmers University of Technology (Sweden) continued the investigation of the methods that allow ex-situ preparation of double-side coated $Tl_2Ba_2CaCu_2O_8$ films on $YAlO_3$, $Y-ZrO_2$ and $LaAlO_3$ substrates. He continued the investigations on the properties and applications of artificial grain-boundary Josephson junctions on bicrystal substrates as well [31-32]. The possibilities to vary the junction parameters by doping the boundary with different elements were studied.

V. Tsaneva developed a technology for fabrication of 2-D Josephson junction arrays in collaboration with Neuchatel University – Switzerland [33].

Investigation of microwave properties of YBCO films: The microwave measurements of high-temperature superconducting samples allow obtaining valuable information about the crystalline properties (granularity) of the sample, the concentrations of the superconducting and normal charge carriers and the dynamics of the magnetic vortices. For this reason, the *Superconductivity and cryoelectronics laboratory* included into its plane of works the investigations of microwave properties of high-temperature superconducting materials. These results of the microwave measurement were used for the obtaining of information about the internal physical parameters of high-temperature superconducting materials (such as microwave conductivity of high-temperature superconducting films, magnetic penetration depth, pinning and dynamic characteristics of magnetic vortex in high-temperature superconducting materials) [34-39].

Analysis was performed of a quasi-TEM wave in a parallel plate resonator (PPR) of arbitrary planar shapes. This enabled to apply the PPR technique for measurement of the surface impedance of thin YBCO films and bulk materials with arbitrary shape when they have one planar surface. On the basis of such conducted investigations, the fixtures with different configurations were elaborated and prepared for studying of the surface impedance of YBCO thin film and bulk materials

Set ups, elaborated for contactless investigation of direct current, low frequency alternating current and microwave properties of high-temperature superconducting materials were especially useful for developing of technology for obtaining of high quality HTS thin film in the laboratory.

Developing of HTS microwave resonators: Microwave losses of high-temperature superconducting thin film materials (expressed by the terms of the surface resistance) are significantly smaller than losses of good conducting metals (Cu, Au, Ag) at $T \leq 77^\circ\text{K}$ and frequencies up to ~ 100 GHz. This allows applying of high-temperature superconducting films for constructing high quality microwave resonators, filters and other microwave elements.

In the *Superconductivity and Cryoelectronics Laboratory*, the microstrip and co-planar resonators of different configurations (linear, ring, meander, circular, elliptic) were prepared

from high-temperature superconducting YBCO thin films (Fig. 4) [34-35, 38]. Quality factor of such microstrip resonators reached up to 10000 at 77°K. A 4 GHz oscillator stabilized by a high-temperature superconducting microstrip resonator was implemented and tested [39]. Parameters of such device strongly exceeded those of Au-resonator stabilized oscillators.

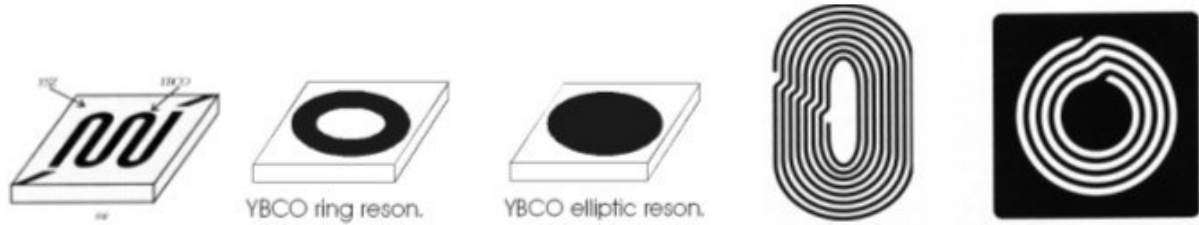


Fig. 4 HTS YBCO thin film microstrip resonators of meander, ring, disc and spiral types

Developing of tunable HTS thin film resonators: The magnetic field effect on the parameters of HTS microstrip line resonators with a ferrite thin film component has investigated. The results obtained were applied for elaboration and analysis of magnetically-tunable high-temperature superconducting resonators of different configuration with the ferrite components [38, 40] (Fig. 5)

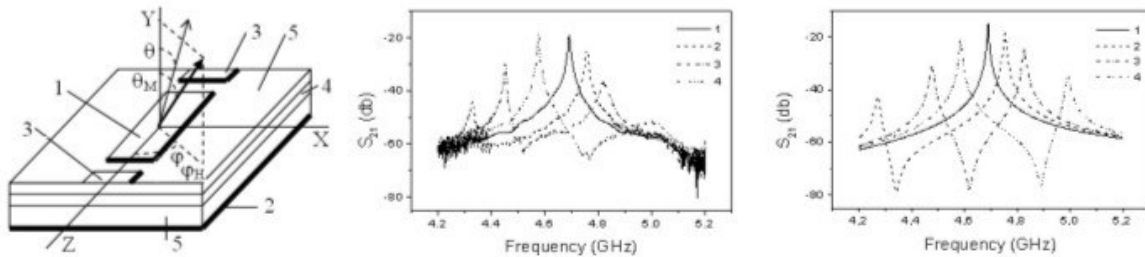


Fig. 5 Transmission characteristics of a HTS YBCO microstrip resonator 2 (left) measured at 77°K (center) [40]. Induction of the magnetic field is $\mu_0 H_e = 0, 77, 85, 93$ mT (curves 1- 4, respectively). Results of modeling (with the parameters $Q_0=1500, f_0=4.68$ GHz, $K_0=0.3, N_1/Q_0=0.002, \alpha=0.0015, \mu_0 M=235$ mT) are shown at right

Co-planar transmission lines were fabricated from YBCO/Ba_xSr_{1-x}TiO₃ bilayers deposited on LAO substrates. Ferroelectric layer [41] in CPW increased and allowed tuning of the effective dielectric constant and the delay time of the pulses. It was shown that an advantage of using high-temperature superconductivity electrodes at 77°K and $f=3 - 10$ GHz could be obtained if the ferroelectric layer thickness was less than 100 nm [42].

Properties of LSMO and YBCO/LSMO structures: The EPR and FMR absorption spectra [43] and the magnetic domain structure [44] of La_{0.7}Sr_{0.3}MnO₃ films prepared on LaAlO₃ (LAO) were investigated. The behavior of the resonance spectrum, observed around the Curie temperature, was interpreted as due to a parallel existence of the microdomains with the paramagnetic and ferromagnetic states of the electron system [43]. Microwave surface impedance, absorption, transmission and reflection coefficients of tangentially magnetized La_{0.7}Sr_{0.3}MnO₃ films [45] and double layer high-temperature superconductivity YBa₂Cu₃O₇ (YBCO)/(FM) La_{0.7}Sr_{0.3}MnO₃ (LSMO) structures [46] with respect to the electromagnetic wave, incident perpendicularly, were analyzed. It was shown that in double layer YBCO/LSMO structures these parameters were characterized with the peculiarities caused by the superconducting transition and the ferromagnetic resonance (FMR). A

dependence of the surface resistance of the structure on the external magnetic field strength can be observed in YBCO/LSMO structures at $T < T_C$ due to the ferromagnetic resonance effect. A contactless method for investigation of the spin –injection effect on the critical current density of high-temperature superconducting film in HTS/FM structures was proposed and realized in a YBCO/LSMO double layer structure [47].

Conclusions

The *Superconducting and Cryoelectronics Laboratory* of the Institute of Electronics BAS has formed in 1989. During these more than 20 years, the laboratory successfully developed the magnetron sputtering technology for preparing of thin films (with a thickness from several teens to several hundred nanometers) and layered structures of modern and attractive materials such as high-temperature superconductors, magnetoresistive ferromagnetic manganites and different buffer layers. Investigation of original physical properties and application possibilities of these thin films was a main field of activity of the laboratory in this period. The scientific problems, in which the team of the laboratory is involved, remain actual and attractive nowadays as well because the new thin film materials and structures are needed in electronics and spintronics.

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