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Inhomogeneities in $YBa_2Cu_3O_7$ thin films with reduced thickness

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ABSTRACT

Morphology and physical properties of mono- ($YBa_2Cu_3O_7$) and multilayered ($YBa_2Cu_3O_7$/$SmBa_2Cu_3O_7$/$YBa_2Cu_3O_7$) superconducting thin films with thickness ranging from 90 nm to 28 nm have been investigated. For both types of samples, the superconducting properties degraded with reduction of film thickness. Structural and electromagnetic properties were visualized through scanning electron microscopy and magneto-optical imaging, respectively, and revealed high level of inhomogeneity for thinner (<58 nm) samples. However, samples with thickness above 58 nm showed enhanced homogeneity, which explains better superconducting characteristics observed in these films. Results of this work demonstrate that multilayering approach performed by conventional laser ablation results in degradation of superconducting properties in films with thickness below 90 nm, although has positive impact on morphology of these films, which is crucial for device fabrication process.

Recently, the ultimate sensitivity for low light level systems has been provided by a new class of nanowire Superconducting Single Photon Detector (SSPD) [1,2]. These devices outperform other superconducting photon counters and are significantly better than commercially available semiconducting Si and InGaAs avalanche photodiodes [3]. To date, single photon sensitive detectors have been based on Nb and NbN superconducting thin films operating at 2.6 K or 4 K [4,5]. There would be operational advantages in utilizing $YBa_2Cu_3O_7$ (YBCO) superconducting thin films operating up to 77 K, which would allow substantially relaxed (by modern cryocoolers) cooling requirements. However, for the potential to be realized, there is a need to develop methods of preparing nanoscale YBCO structures with a high degree of structural perfection.

YBCO films of various thicknesses have been previously extensively investigated [6–9]. Superconductivity in the ultrathin films has shown to be present for thicknesses as small as 1 unit cell [7], but with the critical parameters which are much less than those on thicker (up to micrometer size) films [9]. However, several preceding studied demonstrated that multilayering approach (i.e. alternation of YBCO layers with other superconducting $NdBa_2Cu_3O_7$ [10–12]) or non-superconducting ($PrBa_2Cu_3O_7$ [7], Nd_2CuO_4 [9], CeO_2 [13]) materials drastically improve superconducting properties and morphology of final YBCO film. In spite of relatively thick (up to 1 μm) samples investigated, this approach may be viable for improving superconducting properties of YBCO films at smaller (a few nm) thickness [14].

Being attracted by the idea of possible fabrication of SSPD from YBCO films [15,16], we have initiated study of structural and electromagnetic functionalities of laser ablated YBCO thin films with reduced (from optimal 250–300 nm for our technological process down to 28 nm) thickness. We have also investigated effect of multilayering approach on properties of YBCO films at the thickness down to 44 nm.

The epitaxial, c-axis oriented monolayer YBCO and multilayered YBCO/SmBCO/YBCO (where SmBCO denotes $SmBa_2Cu_3O_7$) thin films were made by pulsed laser deposition using a 248 nm wavelength, KrF eximer laser on (001) $SrTiO_3$ (STO) substrate. A substrate temperature of 780°C and O_2 pressure of 300 mbar were kept during deposition. Bulk superconducting targets (YBCO or SmBCO) were ablated for different lengths of time in order to fabricate films with different thicknesses (Table 1). The frequency at which the laser was incident on the targets was 1 Hz for all samples. After deposition, films were annealed at 400 °C for 1 h at 1 atm of oxygen. The multilayered films were deposited with equal thickness layers of YBCO, SmBCO, and YBCO in this particular order.

Superconducting properties (critical temperature, magnetic moment) of the films were investigated in the Magnetic Property Measurement System (MPMS, Quantum Design). Distribution of magnetic flux in the films was studied by Magneto-Optical Imaging (MOI) at a temperature of 10 K. Images were acquired by a computer-controlled CCD camera with the magnetic field applied perpendicular to the sample surface. The morphology of the films was studied with the help of the Scanning Electron Microscope (SEM). The thickness measurements were carried out using a Dektak 6 M Stylus Profiler. The errors given in Table 1 are the 95% confidence limits (determined by twice the standard deviation) of the 5 measurements taken for each sample.

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Table 1
Samples investigated in this work.

Composition	Deposition time (s)	Thickness, nm	Abbreviation
YBCO	500	90 ± 14	Y90
YBCO	350	60 ± 13	Y60
YBCO	250	28 ± 8	Y28
YBCO/SmBCO/YBCO	500	90 ± 15	Y/Sm90
YBCO/SmBCO/YBCO	350	58 ± 7	Y/Sm58
YBCO/SmBCO/YBCO	250	44 ± 7	Y/Sm44

The surface morphology of the films studied is presented in Fig. 1. As can be seen, the Y28 sample has large cavities on the surface of the film (shown by arrows in Fig. 1a). This implies that the amount of ablated particles was insufficient to achieve full covering of the substrate area (5×5 mm^2). As the deposition time (and thus the thickness of the film) increases, more material arrives; thus, the film continuously covers the surface of the substrate, and becomes notably smoother (Fig. 1b and c).

The Y/Sm44 sample shows the most inhomogeneous morphology among all samples studied (Fig. 1d). Note, the thickness of each layer in this sample is as thin as ~14 nm. We speculate that the first YBCO layer at this thickness consists of many separated islands. This growth mode likely affects growth of the following SmBCO and YBCO layers, and results in observed high level of inhomogeneity and roughness of the final film. However, as the thickness of each individual layer increases, the morphology of the films improves (Fig. 1e and f).

There are no large voids on the surface of $\leqslant$90 nm thick films in contrast to thicker ($\geqslant$250 nm) YBCO films [10]. Indeed, initially strong adatom-substrate bonding results in layer-by-layer growth of the YBCO film on STO substrate up to the thickness of a few unit cells (~6 nm), as confirmed by the Reflection High Energy Electron Diffraction studies [6]. With increasing film thickness up to ~9–19 nm, adatom-adatom bonding prevails leading to the switch from two-dimensional (or layer-by-layer) to three-dimensional (or island) growth occurs [8]. Thus, all samples studied (having thickness in the range 28–90 nm) are grown in a three-dimentional mode, i.e. with increasing the film thickness YBCO islands become large in size and coalesce. The latter results in appearance of voids in YBCO samples. Note, no voids formed in Y28 sample. They are started to be seen at the thickness of 60 nm and have the size of about 10 nm (shown by arrows in Fig. 1b). When the thickness of YBCO film reaches 90 nm, the voids have size in the range from 10 nm to 350 nm (Fig. 1c).

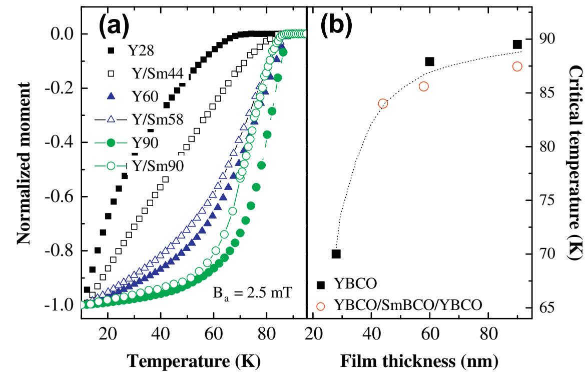

Fig. 2. (a) Normalized magnetic moment as a function of temperature for the samples studied. (b) Critical temperature as a function of film thickness. (The line is a guide for eyes only.)

In contrast, there were no voids obtained in the multilayered films at any thicknesses (Fig. 1d–f). It is likely due to interrupted growth of island's height caused by multilayering. The improved surface morphology of Y/Sm58 and Y/Sm90 films is well suited for structuring a continuous meander line required for fabrication of SSPD device.

Fig. 2a shows magnetic moment of the films as a function of temperature, from which the onset critical temperature values (T_c) are determined and plotted against the film thickness in Fig. 2b. Thinner samples (Y28 and Y/Sm44) have broad T_c transition, which is consistent with highly inhomogeneous structure obtained in these films (Fig. 1a and b), and likely poor crystallinity of YBCO layers in these

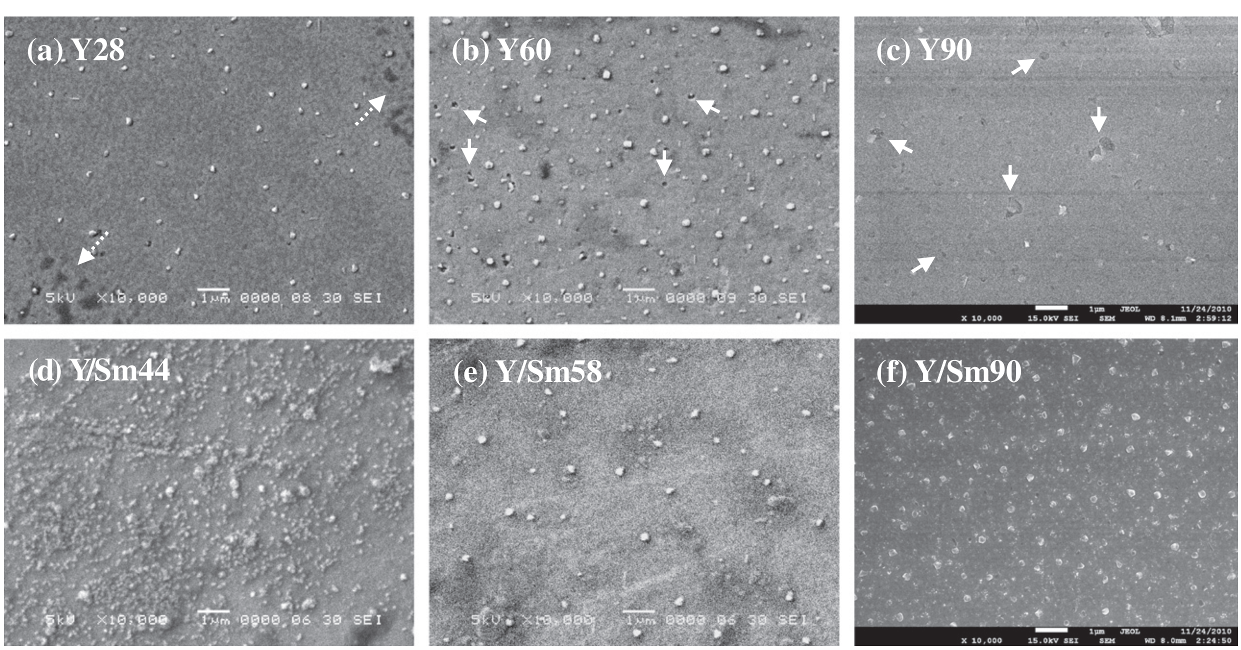

Fig. 1. Morphology of the thin films studied. Images (a–c) and (d–f) correspond to the different thickness of monolayer and multilayered films, respectively. Broken arrows in (a) point to areas with large cavities. Solid arrows in (b) and (c) show some voids formed in 60 nm and 90 nm thick YBCO structures.