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Surface planarization and masked ion-beam structuring of $YBa_2Cu_3O_7$ thin films

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ABSTRACT

Surface planarization and masked ion-beam structuring (MIBS) of high-T_c superconducting (HTS) $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films grown by pulsed-laser deposition (PLD) method is reported. Chemical–mechanical polishing, plasma etching, and oxygen annealing of YBCO films strongly reduce the particulate density (~$10^{-2}\times$) and surface roughness (~$10^{-1}\times$) of as-grown PLD layers. The resistivity, critical temperature $T_c \approx 90$ K and critical current density J_c (77 K)>1 MA/cm^2 of films are not deteriorated by the planarization procedure. The YBCO films are modified and patterned by irradiation with He^+ ions of 75 keV energy. Superconducting tracks patterned by MIBS without removal of HTS material and, for comparison, by wet-chemical etching show same T_c and $J_c(T)$ values. Different micro- and nano-patterns are produced in parallel on planarized films. The size of irradiated pattern depends on the mask employed for beam shaping and features smaller than 70 nm are achieved.

1. Introduction

Oxidic perovskite materials reveal a plethora of dielectric, electric, magnetic, and (multi)ferroic phenomena. Thin films and multi-layers of such materials show additional phenomena as, for example, the formation of a quasi two-dimensional electron gas at the interface of insulating oxides [1,2], the correlation of defects and pinning centers in high temperature superconducting cuprates [3], and the enhanced polarization in asymmetric ferroelectrics [4]. The investigation of these effects requires the fabrication of high-quality thin film samples and the patterning of samples on the micrometer and nanometer scale [5]. Pulsed-laser deposition (PLD) is a versatile and frequently used technique to grow epitaxial oxide layers with controlled composition and lattice orientation [6]. The nucleation of films, the growth of multi-layers and the termination of sample surfaces can be monitored during the PLD process by reflection high energy electron diffraction [5,7]. High-temperature superconducting (HTS) thin films grown by PLD show high critical temperature T_c and critical current density $J_c(T, B)$ and are promising for the development of electronic devices. Among the advantages of HTS materials like $YBa_2Cu_3O_{7-\delta}$ (YBCO) is the strongly reduced dissipation of electro-magnetic energy at reduced temperature as compared to conventional metals. The reduced thermal loading gets relevant when the size of functional structures in electronic devices is reduced to the micrometer and nanometer range and heat conduction becomes one of the critical design issues.

The patterning of oxide thin films by photolithography and etching produces micrometer sized structures, typically. Nano-patterning of films requires advanced technology such as focused ion-beam lithography [8,9]. HTS films grown by PLD have smooth surfaces together with droplets, particulates and outgrowths that deteriorate the surface quality. The number density of such nano- and micro-particles is around 10^4/mm^2 for YBCO films [10] and $Bi_2Sr_2CaCu_2O_{8+\delta}$ films [11] deposited under optimized conditions. The surface irregularities are a severe obstacle for nano-patterning of layers and different techniques have been employed to suppress the formation of those particles, like off-axis PLD, shadow mask PLD, plasma assisted deposition, crossed-beam deposition, and dual-laser ablation deposition. However, these techniques required the optimization of additional experimental parameters to produce samples of comparable homogeneity and quality. The removal of particles after film deposition is an alternative approach for surface planarization. Chemical–mechanical polishing (CMP) enables to smoothen metal, oxides and semiconductor thin films and wafers [12]. The method is employed in semiconductor, microelectronics and micro-systems technology to produce surfaces with roughness in the Å range. The application of this method to functional oxide layers has been discussed in references [13,14], for example.

Here, we report on the surface planarization of pulsed-laser deposited YBCO films by CMP technique. For micro- and nano-patterning of

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planarized films over a larger sample area masked ion-beam structuring (MIBS) technique was employed. The structural properties of YBCO films were investigated by X-ray diffraction (XRD, Cu Kα), scanning electron microscopy (SEM), atomic force microscopy (AFM), optical microscopy, and Rutherford backscattering (RBS). The electrical and superconducting properties of film samples were characterized by electrical transport and conductive AFM (c-AFM) measurements.

2. Experimental details

Epitaxial YBCO thin films were grown on (001) MgO single crystal substrates by PLD. Substrate surface area was 25 mm^2 and substrate thickness was 1 mm, typically. Two identical YBCO film samples were fabricated in each PLD run. The thickness of as-grown YBCO layers was 230 nm for 6000 pulses of a KrF UV excimer laser at a laser fluence of 3.2 J/cm^2 [15]. Surface planarization of as-prepared YBCO thin films was achieved by a three-step process consisting of CMP, plasma etching, and thermal annealing [15]. The films were polished by using a paste containing diamond particles of 100 nm diameter (Kemet company), ethanol, and a developer (Shipley MF-319) and a commercial polishing equipment [16]. In CMP of YBCO films mechanical abrasion by diamond particles and chemical processes (ph-value of developer was > 13) may have contributed. A dependence of removal rate on ph-value was reported for CMP of ferroelectric oxide films [13]. After cleaning of polished films in acetone ultrasonic bath, the residues of diamond paste were etched away in a glow discharge in 0.04 mbar argon background. Samples were placed at a distance of 3–5 mm beside a cylindrical metal rod with diameter of 3 cm. The metal rod was biased at a voltage of +1200 V and film samples were kept at ground potential (Pfeiffer Vacuum 500, Leybold Contraster C2000). After plasma etching, the films were annealed in oxygen (temperature 750 °C, pressure 800 mbar) to compensate for oxygen depletion and to annihilate surface defects introduced by polishing and glow discharge cleaning. XRD analysis demonstrated the *c*-axis orientation of both, as-grown and planarized YBCO films. XRD rocking curve measurements of the (005) YBCO reflection revealed the same profile and full-width-half-maximum $\Delta\omega_{FWHM} = 0.90°$ for both types of films.

Irradiation of YBCO films by He^+ ions of 75 keV energy was performed by means of two different ion beam systems. For homogeneous irradiation of large-area sample surfaces an ion implanter (High Voltage Engineering Europa B.V.) with rapid lateral beam scanning and large exposure field (3×3 cm^2) was used. For MIBS patterning on smaller sample area (~1 mm^2) a Van de Graaff accelerator (AN700 system) was used. Here, the ion beam current was $I = 1 \pm 0.1$ nA, the spot area at the sample position $A_{spot} = 1.33$ mm^2, and the beam current density $J_B = I/A_{spot} = 75$ nA/cm^2. The irradiation dose applied to all samples was 3.0×10^{15} $ions/cm^2$ corresponding to an exposure time of 106 min. The divergence of the ion beam was ≤ 1 mrad and the angle of incidence was 0°. Film samples were irradiated at temperature $T = 300$ K and at background pressure of 10^{-7} mbar.

Different types of masks were employed for ion beam patterning of YBCO thin films. Photoresist (PR) masks were used for MIBS patterning of superconducting current tracks. PR layers of thickness 1.0 to 1.5 µm were spin coated on the YBCO layers and patterned into tracks of 100 µm width and 1.0 mm length by UV photolithography and resist development. The remaining PR layer served as mask to protect underlying tracks and contact pads against ion irradiation. For micro- and nano-patterning of planarized YBCO films commercial silicon (Si) stencil masks 2 µm in thickness were used. The stencil masks had various apertures ranging from 1.5 µm to 125 nm in size. The narrowest Si lamella separating adjacent mask openings had a width of approximately 70 nm (as determined by SEM). Masks were mounted on a stainless steel plate with 1 mm hole to provide mechanical stability. RBS analysis (350 keV D^+ ion beam) was employed to check for deposition of carbon on YBCO films due to He^+ ion induced cracking of residual C_xH_y molecules in the recipient. RBS spectra of pristine films and of He^+ ion irradiated films were measured with a high resolution detector. From RBS measurements the thickness of layers deposited on YBCO surface was estimated to be less than 7 Å.

For comparison, YBCO films were patterned also by standard photo-lithography and wet-chemical etching. Superconducting film tracks were produced by immersing the YBCO sample for 7 s in 0.2 vol.% HCl aqueous solution at room temperature. The edge sharpness of patterned tracks as revealed by SEM inspection was around 1 µm [17].

Electrical transport measurements of YBCO films were performed by four point method. Circular Au contact pads 0.8 mm in diameter were sputtered on the film surfaces prior to patterning (pad area $A_P \approx 5 \times 10^{-3}$ cm^2). The critical current density was determined from current–voltage curves measured by DC currents and by short current pulses (pulse duration 500 µs, duty cycle 4×10^{-3}). The electric field criterion was 10 µV/cm for DC measurement and 300 µV/cm for pulsed measurement due to higher electromagnetic background and noise level.

3. Results and discussion

YBCO thin films with planarized surface morphology, high T_c and high $J_c(T)$ values were obtained by optimizing the parameters of CMP and plasma treatment. Homogeneous plasma etching of samples up to 10×10 mm^2 in size was achieved for films facing the electrode rod at a tilt angle of approximately 10° between sample surface and rod axis. Samples oriented almost perpendicular to the rod axis (tilt angle 80°) were etched non-uniformly and revealed large elliptically shaped etch pits. The planarization procedure reduced the YBCO layer thickness from 230 nm to approximately 200 nm. Fig. 1 shows SEM images of the surface topography of planarized (Fig. 1a) and of as-grown YBCO

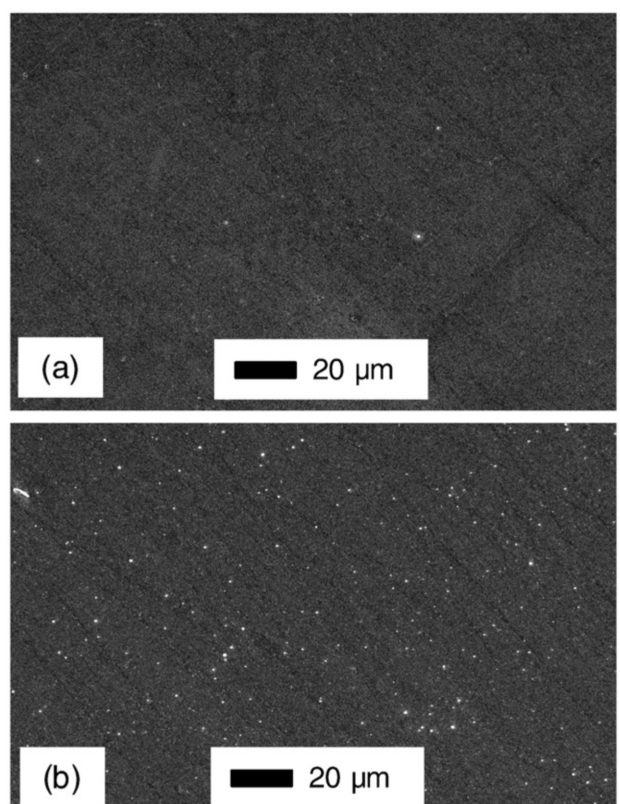

Fig. 1. Scanning electron microscopy of $YBa_2Cu_3O_7$ (YBCO) thin films grown on (001) MgO single crystal substrates by pulsed-laser deposition. Surface morphology of films after *ex situ* planarization treatment (a). Surface of as-grown YBCO films with high density of particulates (b).