Modification and nano-patterning of high-T\textsubscript{c} superconducting thin films by masked ion beam irradiation

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Abstract. Ion irradiation of the high-temperature superconductor (HTS) \textit{YBa}_2\textit{Cu}_3\textit{O}_7\textsubscript{(Y-123)} creates different types of defects depending on ion mass, energy and dose. Irradiation with helium ions of moderate energy (75 keV) primarily creates point defects. We measure in situ the modification of electrical transport properties of Y-123 thin films (thickness 310 nm) during ion irradiation. The He ions penetrate thin films and produce collision cascades with small lateral straggle that allow for patterning of nanostructures in the HTS layer. We present features smaller than 100 nm in size produced by masked ion beam irradiation of Y-123 films. Computer simulations indicate that nano-patterning of Y-123 thin films with 10 nm lateral resolution is achievable.

1. Introduction

High-temperature superconducting (HTS) thin films are promising materials for the development of novel electronic devices. Among the advantages of superconducting materials is the strongly reduced dissipation of electro-magnetic energy at reduced temperature ($T < T_c$) as compared to conventional metals. The reduced thermal loading becomes especially 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.

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The fabrication of novel HTS thin film electronic devices requires advanced technology to pattern HTS materials on the nanometer scale. Focused ion-beam (FIB) lithography is employed to produce planar junctions and heterostructure junctions in HTS materials by scanning the FIB spot across the sample [1]. In masked ion-beam structuring (MIBS) a mask is used for projection patterning of the specimen. MIBS nano-structuring of HTS films using masks in direct contact to the film is reported [2]. Here, we present the measurement of Y-123 film resistance in situ during He+ ion irradiation and the nano-patterning of Y-123 thin films by MIBS using stencil masks at a distance from the sample.

2. Experimental
Epitaxial and c-axis oriented YBa$_2$Cu$_3$O$_7$ (YBCO, Y-123) thin films are grown on (001) MgO single crystal substrates by pulsed-laser deposition (PLD) technique [3]. For electrical characterization of samples, metal contact pads (Au/Ag) are evaporated onto the film surface after plasma cleaning. The YBCO thin films are patterned by photo-lithography and wet-chemical etching producing current tracks that are 100 µm in width and 1.0 mm in length. Electrical measurements are performed by four-point method without external magnetic fields.

For ion irradiation of thin film samples an ion implantation facility (High Voltage company) is employed. The YBCO films are placed with the sample surface perpendicular to the ion beam axis and films are irradiated by He$^+$ ions of 75 keV energy. A stainless steel mask with a 2 mm diameter aperture is placed close to the sample to prevent the metal film contacts from irradiation. For masked ion-beam patterning a thin silicon stencil mask in close proximity to the YBCO film surface is used.

3. Results and discussion
Figure 1 shows the electrical resistance $R$ of a patterned YBCO thin film track at room temperature measured in situ during ion irradiation. The resistance is normalized to the resistance value of the pristine non-irradiated sample ($R_0$).

Figure 1. Variation of YBCO thin film resistance with employed He$^+$ ion fluence (nominal sample temperature $T = 300$ K). The resistance $R$ is measured in situ during He$^+$ irradiation and normalized to the resistance value $R_0$ for the non-irradiated sample. The solid lines are fits to the data.

A strong and non-linear increase of YBCO thin film resistance with employed He$^+$ ion fluence is observed. At a moderate fluence of $7 \times 10^{14}$ cm$^{-2}$, for instance, the sample resistance increases by a
factor of 3. This fluence corresponds to an average dose \( D_{ab} = \frac{1}{ab} \) of approximately one ion per area of the YBCO basal plane which is exposed to the ion beam \((a = 3.82 \text{ Å and } b = 3.88 \text{ Å are YBCO unit cell lattice parameters})\). Measurements of the temperature-dependent resistivity \( \rho(T) \) and critical temperature \( T_c \) of YBCO films performed \textit{ex situ} after irradiation revealed also a strong increase of \( \rho(T) \) and a decrease of \( T_c \) with accumulated ion dose [4]. Non-linear increase of YBCO film resistance with irradiation dose was observed also for hydrogen, fluorine and carbon ions of 200 keV energy [5]. Computer simulations show that \( \text{He}^+ \) ion irradiation primarily creates point defects in YBCO by displacing oxygen atoms from lattice positions to interstitial sites [4]. Displacement of Y, Ba, and Cu atoms is less likely. The resistivity of YBCO depends sensitively on the ordering of oxygen atoms in the unit cell. The creation of interstitial oxygen - lattice vacancy defects by ion irradiation can thus increase the resistivity of the exposed YBCO material.

The measured increase of sample resistance can be described tentatively by modeling the thin film sample as network of resistors. The network is formed by serial and parallel connections of different resistors that represent the pristine and the ion-modified YBCO material. The resistivity of pristine YBCO is \( \rho \) and the resistivity of ion-modified material is described by \( \rho_i = \rho + \Delta \rho \) according to Matthiessen rule [6]. The fraction of modified material \( f \) varies from \( f = 0 \) (pristine sample) to \( f = 1 \) (all sample material modified). The normalized resistance of a network consisting of two resistors in parallel connection and a third resistor in series connection is given by

\[
R/R_0 = 1 + f/r(1-f^z) 
\]

with \( r = \Delta \rho / \rho \). The parameter \( z \) describes the dimensionality of the network. For three-dimensional (3D) and two-dimensional (2D) networks the parameter \( z \) is 2/3 and 1/2, respectively. Parallel (serial) connection of only two resistors is described by \( z = 1 \) \((z = 0)\), for comparison. The parameters \( f, r, \) and \( z \) may depend on the target material, the projectile ions (type and energy of ions), and the experimental arrangement (e.g., ion current density and dose, angle of incidence, sample temperature) [7].

For basic modeling, we assume that the first irradiation induced defect in a YBCO unit cell is most effective in changing the local electric conductivity. The formation of multiple defects in the same unit cell is less probable and the contribution of such defects to the increase of resistivity is neglected therefore. These assumptions lead to an exponential variation of fraction with dose, \( f = 1 - \exp(-D/D_0) \), and a resistivity increase \( r = \text{const} \) that is independent of dose. The solid lines in Figure 1 are fits of the network model resistance to the measured data. Fits performed with different parameters \( z \) show good agreement to the measured resistance data for the range of ion dose investigated. Extrapolation of fit curves to higher dose levels predicts resistance values that significantly depend on the parameter \( z \). A characteristic dose \( D_0 \approx 10.5 \times 10^{14} \text{ cm}^{-2} \) and a resistance increase \( r \approx 585 \) are obtained as fit parameters for the 2D model \((z = 1/2)\). Note that the obtained characteristic dose \( D_0 \) and the average dose \( D_{ab} \) discussed before have similar values.

The \( \text{He}^+ \) ion modification of YBCO can be employed to pattern HTS thin films into superconducting and non-superconducting regions by masking some parts of the films against ion irradiation. The electrical properties (resistivity, critical current density) of YBCO films patterned into tracks by MIBS and, for comparison, by wet-chemical etching are found to be the same [7].

For masked-ion beam nano-patterning of thin film samples the range of penetration and the lateral straggling of projectile ions in the target material are essential parameters. Computer simulations (MARLOWE program) indicate that 75 keV \( \text{He}^+ \) ions can penetrate YBCO layers that are several hundreds of nanometer thick. The calculated lateral straggles of ions is about 10 nm for a 100 nm thick
YBCO film. These results indicate that masked He\(^+\) ion beam patterning of YBCO thin films on the nanometer scale is achievable [4].

An example of a MIBS patterned YBCO thin film is presented in Figure 2. The scanning electron microscopy (SEM) images show the surface of Y-123 thin films that were irradiated with He\(^+\) ions at 75 keV energy and dose of \(3 \times 10^{15}\) cm\(^{-2}\). A thin Si stencil mask was used for patterning by shadowing parts of the film against the beam.

![Figure 2. SEM images of masked ion beam patterned YBa\(_2\)Cu\(_3\)O\(_7\) thin films. The YBCO films were irradiated with He\(^+\) ions using a silicon stencil mask for patterning. Arrows indicate ion-irradiated arrays of nano-dots (a) and of line-shaped patterns (b) according to the Si mask structures.](image)

The SEM images are taken by collecting the electrons back-scattered from the thin film samples. The images show darker contrast (reduced electron yield) for regions that were irradiated by the ion beam (indicated by arrows in red color). Arrays of nano-dots (a) and of line-shaped patterns (b) are produced according to the mask structures. The size of irradiated features at the YBCO film surface is limited mainly by the size of mask structures and the divergence of the incident ion beam [7, 8].

4. Conclusions
Irradiation with light ions at moderate energies (75 keV) can be used to tailor the electrical properties of Y-123 thin films. \textit{In situ} measurements of Y-123 film transport properties during ion irradiation revealed a strong and non-linear increase of film resistance with applied ion dose. Masked ion beam structuring of Y-123 thin films produced different patterns smaller than 100 nm in size.

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