Pulsed Laser Deposition of Patterned Multilayers for HTS Device Fabrication

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Abstract - **Pulsed laser deposition (PLD) has been used to deposit high quality multilayer thin films of Permalloy/Au/YBCO onto (100) oriented substrates of MgO and $SrTiO_3$. These multilayer structures are currently being used to investigate the effect of the injection of polarized electrons on the order parameter of high temperature superconducting (HTS) thin films. To observe this effect, thin films with sharp interfaces (to minimize spin scattering) and low contact resistance will be required. The morphology and structure of the deposited films has been investigated using scanning electron microscopy and x-ray diffraction, respectively. The electrical properties (T_c and J_c (77K, B=0)) were measured for both unpatterned and patterned films. Films were patterned using both wet chemical and ion beam (Ar^+, 1 KeV) techniques. Unpatterned films were characterized inductively as having T_c's > 88 K and J_c's > 1 MA/cm^2. Wet chemical etching resulted in films with slightly reduced T_c's and J_c's ~ 10^3 - 10^4 A/cm^2 however, no reduction in T_c or J_c was observed for dry etching. A lift off procedure using PMMA/Cu has been explored to define smaller (~400 μm) features. Extremely low contact resistance's (< 10^7 Ω cm^2) have been measured for Au films deposited by PLD onto YBCO. The low contact resistance is attributed to the high kinetic of the Au particles. These device structures can be used to develop simple HTS based transistors.**

I. Introduction

There is growing interest in electronic devices based on the principles of 'spin-polarized' transport [1]. This phenomenon utilizes the magnetically polarized electrons present in ferromagnets. We are applying spin-polarized transport in a three terminal geometry in order to control the order parameter in a high temperature superconductor (HTS) thin film. Injection of spin-polarized carriers into an HTS material is thought to reduce the superconducting order parameter over a distance characterized by the spin-diffusion in that material [2],[3]. Controllable weak links in superconducting films by quasiparticle injection schemes has been observed by Langenberg [4],[5] and several others [6]. The advantage for current switching is that gap suppression permits operation at lower power, and therefore, enhanced gain. Fast switching times could be achieved using HTS materials (compared to metals) since the they posses intrinsically short quasiparticle relaxation times.

This paper addresses the fabrication issues in developing spin injection devices in HTS films using pulsed laser deposition (PLD) [7] and standard lithography techniques [8],[9]. Issues such as homogeneity of individual layers and quality of interfaces are critical to the success of the device, since spin-flip scattering can occur, reducing the population of spin-polarized carriers present.

A. YBCO Surface Morpholgy

A critical issue in HTS device fabrication is the surface roughness of the HTS film. Sharp interfaces, free of particulates and surface outgrowths, are required for the sucessful implimentation of the spin injection device, since these defects can act as spin scattering sites. Additonally, the YBCO surface morpholgy has a tremendous effect on the quality of the permalloy magnetization [3]. Although surface outgrowths (e.g., CuO) are common in YBCO thin films [10], adjustment of deposition conditions (substrate tempearture, oxygen deposition pressure, laser fluence and repetition rate) allows minimization of these rough surface features.

B. Contact Resistance at HTS-Normal Metal Interfaces

Deposition of electrodes with low contact resistance is a major issue for HTS device applications as large contact resistance can lead to resistive heating. Several groups have investigated contact resistance to bulk and thin films of YBCO [11]-[13]. The lowest contact resistance are achieved with Au, which has a low bulk resistivity (2.35 μΩ-cm) and forms the most thermodynamically stable interface ($\Delta G > 0$ for reactions of Au with HTS constituents). Using deposition techniques such as e-beam evaporation or sputter deposition, contact resistance's from 10^{-3}-10^{-5} Ω-cm^2 have been measured. PLD offers advantages over conventional physical vapor deposition techniques for low resistance contacts by providing high levels of electronic excitation and variable kinetic energy of the incident atoms [7]. The role of the deposition species excess energy on film growth and interface quality is uncertain, however, *in-situ* contact resistance on the order of 10^{-7} Ω-cm^2 are easily achievable.

C. Patterned Multilayers and 'Top-Down' Spin Injection

Fig. 1 shows the device geometry for the injection of spin polarized current into YBCO. This structure avoids the ambiguity of spin injection through a cross-stripe of the ferromagnetic material (permalloy, $Ni_{0.81}Fe_{0.19}$) where the primary effect occurs across the permalloy-Au-YBCO interface along the long axis of the YBCO/Au line. With this geometry, a uniform population of polarized spins is assumed

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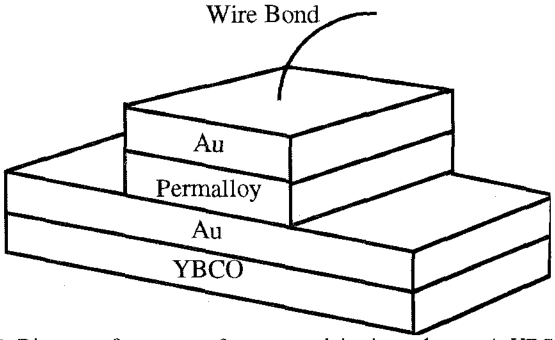

Fig. 1. Diagram of geometry for current injection scheme. A YBCO/Au bilayer is etched to a line and the permalloy/Au bilayer is deposited through a via defined by a PMMA/Cu lift-off mask.

along the surface area defined by the permalloy structure. Here the gold serves two purposes: 1) a diffusion barrier to reduce the loss of O_2 along the YBCO surface and 2) a barrier to avoid the formation of a YBCO-permalloy spin glass phase along the permalloy-YBCO interface [14]. The spin diffusion length has been measured to be on the order of ~1 µm for Au films [15], hence Au thickness from 100-1000 Å should be nearly transparent to the spin-polarized carriers. This geometry also alleviates the need for planerization layers required to keep the material thickness requirements, avoiding circuit breaks and shorts that can occur with overlapping stripes.

II. Experimental

A. PLD of Thin Films

A schematic diagram of the PLD system is shown in Fig. 2. A base pressure of 1×10^{-7} Torr is maintained with a turbo pump (Leybold) and a He cryo-pump (CTI-Cryogenics). Laser ablation is carried out with a KrF excimer laser providing up to 1 J of 248 nm radiation over a 30 ns FWHM pulse (Lambda Phsyik LPX 300). The laser output is focused using a spherical quartz lens (f = 50 cm, D = 10 cm) to provide a fluence of ~10^8 W/cm^2 at the target. Rastering of the focused beam over a rotating 5 cm diameter target is accomplished by placing a computer controlled (Newport MM3000 Motion Controller) mirror prior to the focusing lens.

Substrates of MgO (100) or STO (100), are silver painted onto a stainless steel stage heated by incandescent tungsten bulbs providing uniform substrate temperatures (T_S) up to 900 °C. The system target-substrate distance (d_{TS}) was variable from 6-9 cm. A constant chamber pressure is maintained during deposition by a solonoid controlled leak valve while the turbo pump is throttled. The system is also equipped with a 3 cm Kauffmann ion gun (Commonwealth Scientific) placed ~40 cm from the substrate at an angle of ~30°.

YBCO deposition is carried out at T_S = 750-790°C and $p(O_2)$ = 0.30-0.35 Torr. A quench (rapid or slow) to room temperature is carried out in ~760 Torr of O_2 following deposition. Transport properties (T_c and J_c (77 K, B = 0)) of unpatterned films were determined inductively; crystal

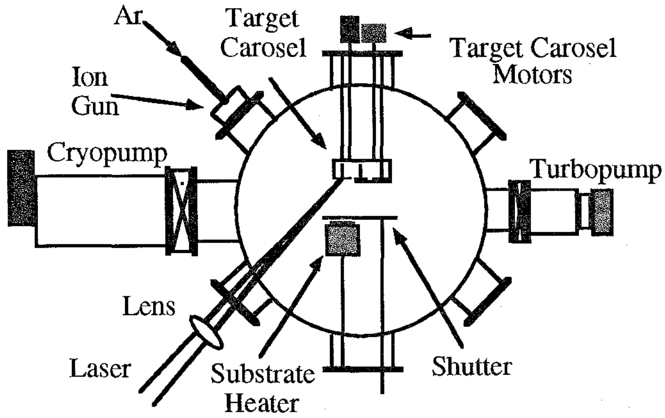

Fig. 2 - Schematic diagram of PLD chamber.

structure by x-ray diffraction (XRD); film surface morphology by scanning electron microscope (SEM). Deposition of metals was carried out over a range of substrate temperatures (T_S) and oxygen pressures (p (O_2)) as indicated in the results section.

B. Lithography

Wet chemical etching of YBCO was carried out using either 1% HCl (aq) or 5% H_3PO_4 (aq) as etchant following pattern definition using AZ1818 positive photoresist and 351 Microposit Developer (1:1 dilution). When starting with a YBCO/Au bilayer film, the YBCO etch step is preceded by a Au etch using a KI/I_2 (aq) solution. Ion etching of YBCO/Au bilayers has been accomplished through masks made from either stainless steel or slices of a MgO substrate (width ~1 mm). Typical etch times are ~ 20 min with a 1 KeV Ar^+ beam. Substrate temperature, monitored through the stainless steel heating stage, is kept at $T_S \leq 40$ °C during the duration of ion etching.

Lift-off masks for deposition of the ferromagnetic and metallization layers consist of a patterned PMMA/Cu bilayer made by spinning on PMMA and baking at 200 °C for 1 hour, followed by deposition of 750 Å of Cu by e-beam evaporation. The lift-off pattern is defined by etching the PMMA/Cu bilayer using AZ1818/351 development followed by: 1) Cu etch using dilute $FeCl_3$ (aq) [8] and 2) PMMA etch using deep UV exposure followed by a soak in methyl isobutyl ketone (MIBK). Prior to the deposition of permalloy, the residual PMMA in exposed areas is removed by exposure to an oxygen plasma. Following deposition, the lift-off mask is removed by soaking in acetone.

III. Results and discussion

A. YBCO Transport Properties

Fig. 3 shows the scatter in transport properties for unpatterned YBCO films grown on MgO (100). The ability of the PLD to make reproducible films is an important aspect since characterization of films tends to create defects and introduce impurities that degrade surfaces and hence, the

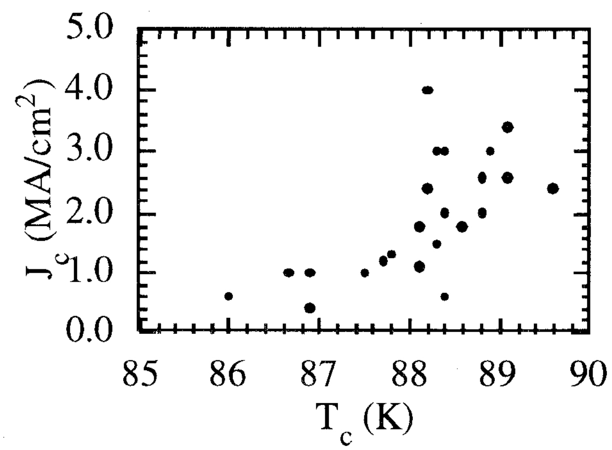

Fig. 3. T_c and J_c (77K, B=0) scatter plot for YBCO thin films grown on MgO (100). PLD parameters: T_s = 750 °C, p = 0.30 Torr O_2, $\Phi = 8 \times 10^7$ W/cm^2, 2500 or 4000 laser shots, slow quench with 1 hour soak in ~ 1 Atm. O_2 at 500 °C.

quality of the interface, as well as the ability to pattern precise structures. Patterning of YBCO or YBCO/Au thin films (1000-4500 Å) to linewidths of 50-400 μm by wet chemical etching has shown, by a four point probe measurement, a slight degradation and a broadening of T_c accompanied by a reduction in J_c by several orders of magnitude. The reduction in J_c is attributed to penetration of the etchant into the interior of the YBCO line along grain boundaries. Post patterning annealing in flowing O_2 at 500 °C has had no effect on the transport properties.

Ion milling with an 1 KeV Ar^+ beam, a 'dry' etching process, has proven to be more successful for the etching of thin YBCO/Au bilayers grown on STO. Fig. 4 shows a I vs. V curve for a YBCO/Au (750/500 Å) bilayer ion etched to 2 mm line followed by PLD of thick (3000 Å) Au cross-stripes (electrodes) through a shadow mask. The transition to superconductivity occurs at ~88 K and is sharp (ΔT_c = 0.3 K) and $J_c = 9 \times 10^5$ A/cm^2 measured at each device electrode pair position. Subsequent milling will utilize masks defined by a PMMA/Cu lift-off in order to define more precise features.

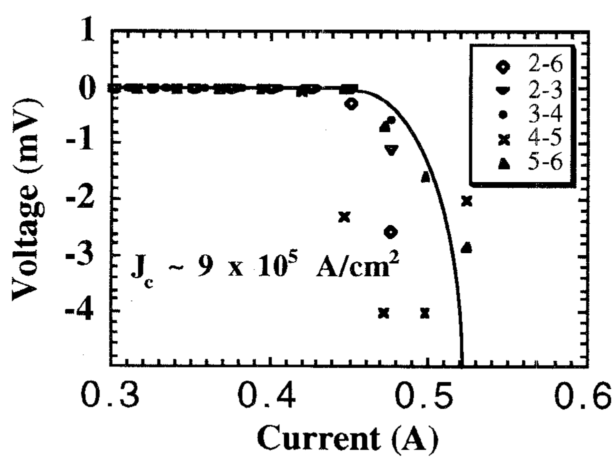

Fig. 4. Transport Properties on YBCO/Au bilayer (750/500 Å) grown on STO (100) and ion milled to a 2 mm wide line. Shown is voltage vs. current curves along each device electrode (Au cross stripe) pair (numbered 2-6).

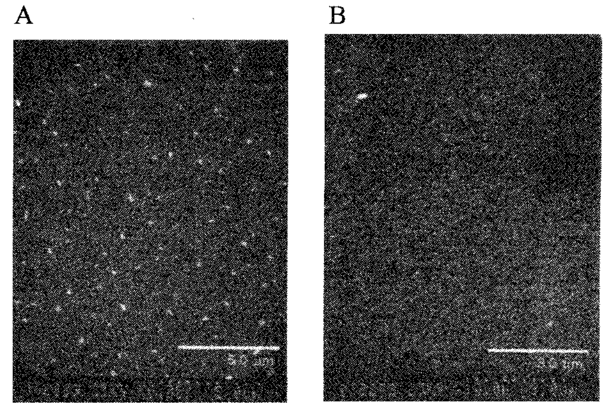

Fig. 5. SEM of YBCO surface morpholgy at different PLD target-substrate distances (d_{TS}). A) d_{TS} = 7.50 cm B) d_{TS} = 6.25 cm

B. YBCO Surface Morphology

Fig. 5 shows the change of YBCO surface morphology as a function of decreasing target-substrate (d_{TS}) distance. For a given d_{TS}, other processing parameters have little effect upon the surface morphology. This effect is attributed to the degree of excess energy contained in the deposited species. At smaller d_{TS} the impinging adatoms have a higher content of excess energy resulting in smoother films.

C. YBCO-Au Contact Resistance

Table 1 lists the relevant PLD parameters and the measured contact resistances for YBCO-normal metal contacts deposited through a metal shadow mask. The contact geometry consists of a circular island of metal of diameter 1.5 mm located at the corners of a square with sides of length 7.5 mm. Values reported less than 1 μΩ-cm^2 are treated as upper limits due to the large cross-sectional area defined by the Au islands. The lowest contact resistances occur for deposition of Au at substrate temperature of 500 °C and no background ambient ($p \leq 10^{-5}$ Torr). These results show that PLD provides lower resistivity (10^{-7} Ω-cm^2) than other physical deposition techniques. In most cases a post annealing process lowers the contact resistance by an order of magnitude.

TABLE 1. CONTACT RESISTANCE AS A FUNCTION OF DEPOSITION CONDITIONS FOR PLD OF AU ON YBCO

Temperature (°C)	Pressure (Torr)	Post Anneal[a] in O_2	Resistance (μΩ-cm^2)
25	$\leq 10^{-5}$ Torr	no	> 1000
25	$\leq 10^{-5}$ Torr	yes	0.30
500	$\leq 10^{-5}$ Torr	no	0.16
500	$\leq 10^{-5}$ Torr	yes	0.90
500	$\leq 10^{-5}$ Torr	no	0.20[b]
500	0.3 Torr O_2	no	1.40
700	0.3 Torr O_2	no	60.7

[a] Post annealing done at T = 600 °C, ~1 Atm O_2
[b] Sputter cleaned with Ar ion beam prior to deposition

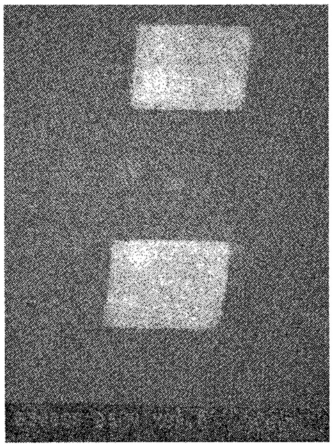

Fig 6. SEM of permalloy deposited on Si through 400 μm x 400 μm via defined by PMMA/Cu lift-off mask.

D. Deposition of Patterned Structures

Fig. 6 shows the result of the deposition of permalloy on silicon substrate through a 400 μm x 400 μm via defined by a PMMA/Cu lift-off mask. Close inspection shows that edges are sharp around the patterned material. Of principle concern was the degradation of the PMMA/Cu lift-off mask during the energetic PLD process.

IV. Conclusions

High quality thin films and multilayers of permalloy/Au/YBCO have been deposited using PLD onto substrates of MgO and $SrTiO_3$. These structures are currently being used to investigate the effect of spin polarized electrons on the superconducting order parameter. The injection of spin polarized carriers into a superconducting thin films offers the possibility of creating a local reduction in the superconducting order parameter which could be used to develop an HTS based transistor. For this device, thin films with sharp interfaces (to minimize spin scattering) and low contact resistance will be required.

To study the effect of polarized electrons on the properties of high temperature superconductors, HTS lines (50 - 400 μm) have been defined both by wet chemical etching and by dry, ion beam processing (Ar^+, 1 keV). As-deposited, unpatterned films were characterized inductively as having T_c's > 88 K and J_c's > 1 MA/cm^2. Wet chemical etching resulted in films with slightly reduced T_c's and J_c's ~ 10^3 - 10^4 A/cm^2 however, no reduction in T_c of J_c was observed for dry etching. Masks for dry etching consisted of stainless steel shadow masks to define relatively coarse structures. A lift off procedure using PMMA/Cu has been explored to define smaller (~400 μm) features. Extremely low contact resistance's (< 10^7 Ω–cm^2) have been measured for Au films deposited by PLD onto YBCO. Using these structures, a reduction in the superconducting order parameter for HTS thin films has, in some cases, been observed for the first time [3].

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