Supercond. Sci. Technol. **11** (1998) 270–272. Printed in the UK PII: S0953-2048(98)87343-0

Preparation of smooth $YBa_2Cu_3O_y$ and $EuBa_2Cu_3O_y$ films by two-beam excimer laser deposition

T Schauer†, L Weber†, J Hafner†, O Kus†, E V Pechen‡, A V Varlashkin‡, T Kaiser§ and K F Renk†

† Institut für Angewandte und Experimentelle Physik, Universität Regensburg, Universitätsstrasse 31, 93040 Regensburg, Germany
‡ Lebedev Physics Institute, RAS, Leninskii Prospeckt 53, 117924 Moscow, Russia
§ Fachbereich Physik, Universität Wuppertal, Gauss-Strasse 20, 42097 Wuppertal, Germany

Received 4 September 1997

Abstract. A laser deposition arrangement for the preparation of smooth $YBa_2Cu_3O_y$ and $EuBa_2Cu_3O_y$ films is presented. An excimer laser beam is split by a knife-edge prism into two beams, which are focused onto two rotating targets oriented perpendicular to each other. The two laser-induced molecular streams collide and form a new stream containing mainly light molecules and almost no droplets. The molecules from this stream deposit on a substrate that is placed in the shadow of the original molecular streams. The use of copper-poor targets prevented the occurrence of precipitates.

1. Introduction

The pulsed laser deposition (PLD) technique has been found to be suitable for the preparation of semiconductor [1], metallic [2], superconductor [3] or high-T_C superconductor [4] thin films. Usually the films contain droplets and other macroparticles [5]. Several methods have been applied to reduce the numbers of droplets and particles, for example PLD in off-axis geometry [6], placing the substrate in a tube heater oriented perpendicular to the target surface, and PLD with filtration of the ablation plasma with a velocity filter [7]. Another method uses the crossed-flux technique [8, 9], with two laser-induced molecular streams colliding and forming a new stream directed onto the substrate which is placed in the shadow area of the two original molecular streams. Compared with other methods, this process has some advantages. For example, it is possible to view the laser-induced molecular streams directly to control the deposition. It is also possible to install *in situ* characterization methods (e.g. RHEED, reflection high-energy electron diffraction). Up to now Nd–YAG lasers have been used for crossed-flux deposition. It is, however, well known that UV lasers are more suitable for the preparation of high-T_C films [10].

Here we report on the preparation of high-quality $YBa_2Cu_3O_y$ and $EuBa_2Cu_3O_y$ films on MgO (001) with a modified crossed-flux technique using an excimer laser.

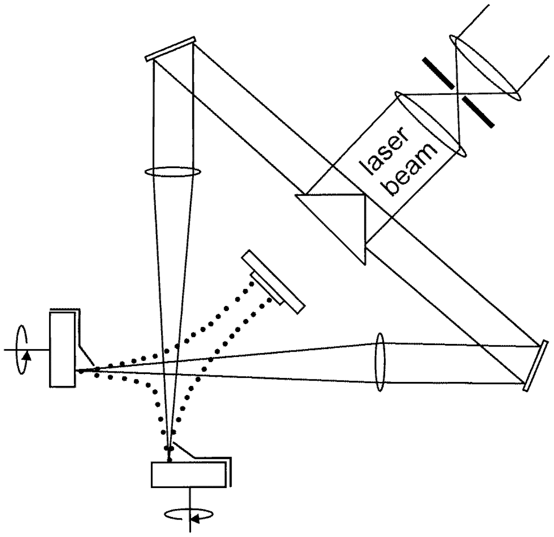

Figure 1. Experimental set-up for two-beam excimer laser ablation.

2. Experiment

The beam of an excimer laser (wavelength 248 nm, pulse duration 20 ns, pulse energy 600 mJ, repetition rate 10 Hz, beam size 32 × 15 mm^2) was split into two beams (size 15 × 16 mm^2) (figure 1) with a knife-edge prism (right-angled

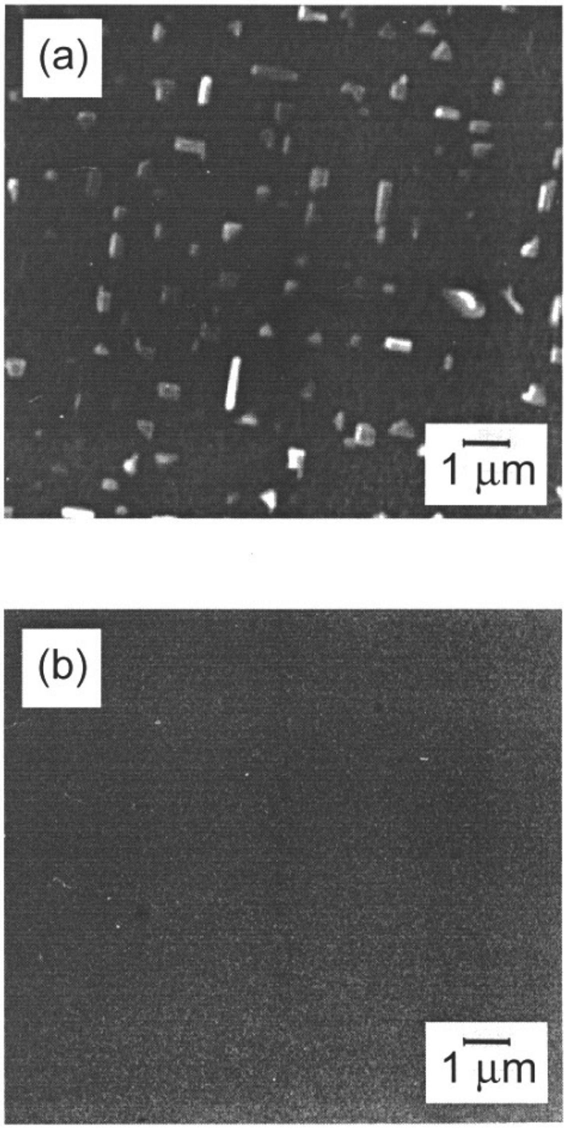

Figure 2. Scanning electron microscope image of 100 nm thick $YBa_2Cu_3O_y$ films, ablated from (*a*) $YBa_2Cu_3O_y$ and (*b*) $YBa_2Cu_{2.6}O_y$ targets.

faces with a high-reflection coating for 248 nm radiation at an angle of incidence of 45°). The beams were reflected by two mirrors, coated with a high-reflection coating (for an angle of incidence of 22.5°). Finally the beams were focused by two lenses (with low-reflection coatings) onto two rotating disc-shaped targets of $YBa_2Cu_{3-x}O_y$ or $EuBa_2Cu_{3-x}O_y$ ($x = 0$ to 0.5), oriented perpendicular to each other. The distance between substrate and crossing point of the laser-induced molecular streams was about 35 mm. The pulse energy density at the target surfaces was about 5 J cm^{-2}. The targets were prepared by the ceramic method [11] using Y_2O_3, $BaCO_3$ and CuO powders.

Due to collisions of ions, the two laser-induced molecular streams are deflected from their initial directions and are able to reach the substrate (indicated by dotted curves in figure 1). The metallic diaphragms prevent heavier particles, which are not deflected by the other stream, from directly hitting the substrate. The substrate is placed in the shadow region of the two molecular streams.

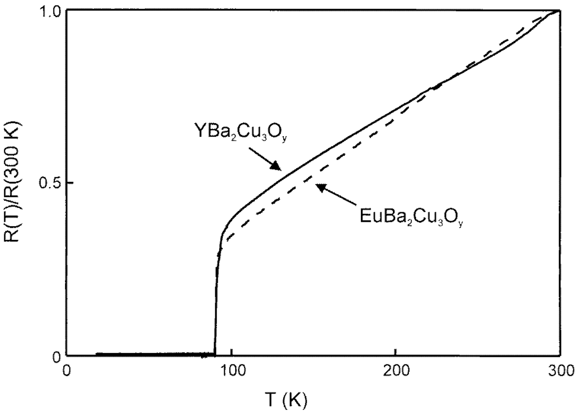

Figure 3. Temperature dependence of the resistance of 150 nm thick $YBa_2Cu_3O_y$ and $EuBa_2Cu_3O_y$ films.

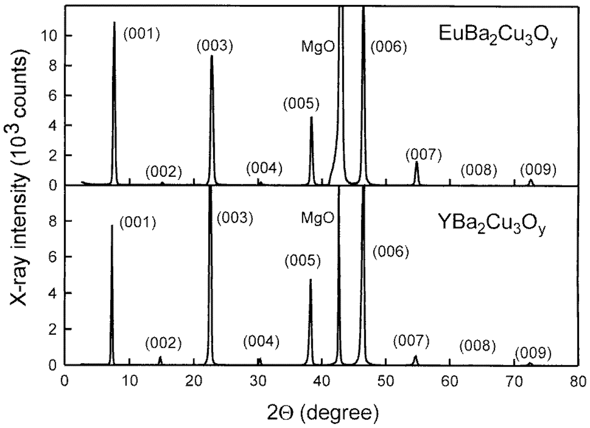

Figure 4. XRD θ–2θ patterns of 150 nm thick $YBa_2Cu_3O_y$ and $EuBa_2Cu_3O_y$ films.

The films were deposited on MgO (001) substrates (size 10×10 mm^2) heated up to 730 °C in an atmosphere of about 30 Pa oxygen. The deposition rate was 110 Å min^{-1}.

We prepared films with thicknesses between 20 and 250 nm, the thickness being determined with a surface profiler and low-angle x-ray diffraction. The critical temperature was determined with mutual inductive and four-probe a.c. measurements. The surface resistance was measured using a cavity resonator at 87 GHz. Structural and crystallographic properties were investigated by x-ray diffraction (XRD) analysis using a four-circle diffractometer and Cu K_α radiation ($\lambda = 1.5406$ Å).

3. Results and discussion

The $YBa_2Cu_3O_y$ and $EuBa_2Cu_3O_y$ films appeared optically mirrorlike over the whole sample area (10×10 mm^2). Scanning electron microscope investigations showed that the surfaces were almost free from droplets with a size

>1 μm and the density of smaller droplets was less than 10^3 cm^{-2}; large clusters, which are present in both plasma streams in small concentrations, are hardly deflected by the light particles, and therefore do not reach the substrate.

However, we observed a high concentration of precipitates on films prepared with $YBa_2Cu_3O_y$ targets (figure 2(*a*)), consisting mostly of copper oxide as determined by EDX (energy dispersive x-ray analysis) investigations. After reducing the copper content of the targets to 2.6 (e.g. using $YBa_2Cu_{2.6}O_y$) the surfaces of our films contained almost no precipitates (figure 2(*b*)).

The temperature dependence of the resistance R of 150 nm thick films is shown in figure 3. Superconductivity occurs for $YBa_2Cu_3O_y$ at 89.7 K and for $EuBa_2Cu_3O_y$ at 93.3 K. The resistive transition width (10%–90%) was about 1 K for both materials. The relation between resistance at room temperature and at 100 K was in the range of 2.5 to 2.9. The films had surface resistances (at 87 GHz) of 3 to 5 mΩ at 4.2 K and about 60 mΩ at 77 K.

XRD θ–2θ patterns of 150 nm thick $YBa_2Cu_3O_y$ and $EuBa_2Cu_3O_y$ films are shown in figure 4. All reflections (except the substrate reflex) can be assigned to the (00l) reflections of the film material, no reflexes of other orientations (e.g. a-axis) or of other phases are observed. This indicates single-phase films and a growth direction of the films with the c-axis perpendicular to the substrate surface. Rocking curves, namely θ scans of the (005) reflection, had a full width at half-maximum of less than 0.4°, indicating a high degree of c-axis orientation.

Using the (018) reflection of $YBa_2Cu_3O_y$ or $EuBa_2Cu_3O_y$ and rotating the film about the surface normal (Φ scan) we found sharp peaks with a separation of 90° and a full width at half-maximum of about 6° indicating a high orientation of the films in the a/b plane. A comparison of these reflections with those of the MgO(001) substrate ((024) reflex) showed that the [100] axis of the films were aligned parallel to the [100] axis of the substrate.

4. Conclusion

In conclusion, our results demonstrate that the two-beam excimer-laser deposition arrangement is an effective and convenient method for preparation of almost droplet-free thin films. Reduction of Cu content in the targets avoids the creation of precipitates.

Acknowledgments

The work was supported by the Bayerische Forschungsstiftung through the Bayerische Forschungsverbund Hochtemperatur-Supraleiter (FORSUPRA) and the Deutscher Akademischer Austauschdienst (DAAD).

References

[1] Lubben D, Barnett S A, Suzuki K, Gorbatkin S and Greene J E 1985 *J. Vac. Sci. Technol.* B **3** 968

[2] Kools J C S, Nillesen C J C M, Brongersma S H, van de Riet E and Dieleman J 1992 *J. Vac. Sci. Technol.* A **10** 1809

[3] Treece R E, Horwitz J S, Claasen J H and Chrisey D B 1994 *Appl. Phys. Lett.* **65** 2860

[4] Roas B, Schultz L and Endres G 1988 *Appl. Phys. Lett.* **53** 1557

[5] Proyer S, Stangl E, Borz M, Hellebrand B and Bäuerle D 1996 *Physica* C **257** 1

[6] Holzapfel B, Roas B, Schultz L, Bauer P and Saemann-Ischenko G 1992 *Appl. Phys. Lett.* **61** 3178

[7] Pechen E V, Varlashkin A V, Krasnosvobodtsev S I, Brunner B and Renk K F 1995 *Appl. Phys. Lett.* **66** 2292

[8] Golovashkin A I, Ekimov E V, Krasnosvobodtsev S I, Martovitsky V P and Pechen E V 1989 *Physica* C **162–164** 715

[9] Gaponov S, Gavrilov J, Jelinek M, Kluenkov E and Mazo L 1992 *Supercond. Sci. Technol.* **5** 645

[10] Kautek W, Roas B and Schultz L 1990 *Thin Solid Films* **191** 317

[11] Rao C N R, Nagarajan R and Vijayaraghavan R 1993 *Supercond. Sci. Technol.* **6** 1