Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl17

Crystallizations of Amorphous Y-Ba-Cu-O Film Prepared by RF-Sputtering

N. Moriyama [a], K. Nobugai [a], M. Takahashi [a], S. Kikkawa [a] & F. Kanamaru [a]

[a] The Institute of Scientific and Industrial Research, Osaka University, 8-1 Mihogaoka, Ibaraki, Osaka, 567, Japan

Version of record first published: 22 Sep 2006.

To cite this article: N. Moriyama , K. Nobugai , M. Takahashi , S. Kikkawa & F. Kanamaru (1990): Crystallizations of Amorphous Y-Ba-Cu-O Film Prepared by RF-Sputtering, Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics, 184:1, 225-229

To link to this article: http://dx.doi.org/10.1080/00268949008031766

Mol. Cryst. Liq. Cryst., 1990, vol. 184, pp. 225–229
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

CRYSTALLIZATIONS OF AMORPHOUS Y-Ba-Cu-O FILM PREPARED BY RF-SPUTTERING

N. MORIYAMA, K. NOBUGAI, M. TAKAHASHI, S. KIKKAWA AND F. KANAMARU
The Institute of Scientific and Industrial Research, Osaka University, 8-1 Mihogaoka, Ibaraki, Osaka 567, Japan

Abstract Amorphous $YBa_2Cu_3O_y$ itself and $YBa_2Cu_3O_y$ containing F or excess amount CuO films were prepared by rf-sputtering. Film texture such as preferred orientation and superconductivity were investigated on these films crystallized in oxygen flow at various temperatures. Substrate effects were also studied on MgO (100) and $SrTiO_3$ (100) and (110) in relation to applied rf-power on sputter deposition. The addition of foreign elements were effective to lowering the crystallization temperature on MgO substrate. The preferred orientations of YBCO on $SrTiO_3$ changed with the deposition conditions of amorphous film.

INTRODUCTION

Superconducting thin film can be obtained by rf-sputtering on high temperature substrate ($\gtrsim$700°C) as-deposited. Deposition on low temperature substrate gives amorphous film, where we can homogeneously add foreign elements because of the wide compositional variation of amorphous state. Its film texture can be controlled by changing the crystallization conditions. We have investigated crystallizations of amorphous β-Al_2O_3[1] and barium ferrite[2] films. Addition of fluoride to $YBa_2Cu_3O_y$ have been studied, but its role is still ambiguous[3].

EXPERIMENTAL

Amorphous Y-Ba-Cu-O thin film was prepared by rf-sputtering using $YBa_{3.45}Cu_{3.50}O_y$ sintered target (φ80x5mm). Addition of F performed by putting exta small targets (φ10x1mm) containing YF_3, BaF_2 and CuO on the above-mentioned large target. Sputter gas was a mixture of Ar/O_2 = 1 and the pressure was 1.8Pa. The deposition rate was 0.35∿0.55 μm/h. Substrates were MgO and $SrTiO_3$ single crystals. Crystallization was performed in O_2 flow at desired temperatures. ICP emission analysis showed the chemical composition of the films without additives were very close to $YBa_2Cu_3O_y$.

RESULTS and DISCUSSION

Amorphous films with 0.8μm thickness on MgO were annealed in a temperature range of Ta = 920∿1020°C[4]. YBCO crystallized but preferred orientation was not clear in Ta < 950°C. Remarkable preferred orientation of YBCO <001> perpendicular to substrate was observed above Ta ≥ 970°C accompanying a pronounced grain growth. Amorphous film with fluoride on MgO substrate can be crystallized to a mixture of $YBa_2Cu_3O_y$ and BaF_2 at 700°C in oxygen flow for 2hrs. The BaF_2 was not observed on the films heated above 800°C for 2hrs. Fluoride addition enhanced grain growth and preferred orientation of $YBa_2Cu_3O_y$ platy crystal. The film with F heated at 920°C had a strongly preferred orientation of $YBa_2Cu_3O_y$ crystal. $YBa_2Cu_3O_y$ crystals were preferred oriented in the film with F heated at 920°C as well as in the film without F heated at 1020°C. Figure 1 shows X-ray diffractions of fluoride added films heated in oxygen flow at 920°C for various

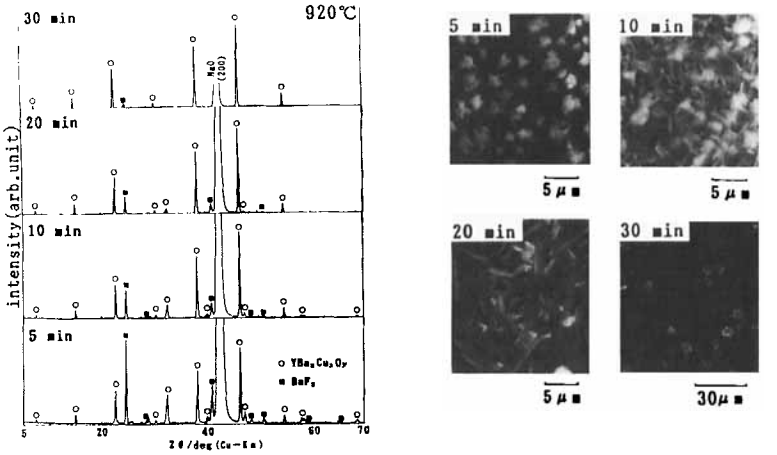

Fig. 1 X-ray diffraction and surface texture of F added $YBa_2Cu_3O_y$ films/MgO with oxygen annealing in various durations.

durations. The film crystallized to a mixture of $YBa_2Cu_3O_y$ and BaF_2 in 5 min heating. The amount of BaF_2 decreased and c-axis preferred orientation was enhanced with an increasing heat duration. SEM observation showed that the YBCO platy crystals grew and the amount of flower-like BaF_2 crystals decreased with an increasing heat duration. EPMA investigation showed that a fluoride amount in the annealed film decreased with an increase of heating temperature and that fluoride was not detected on the films heated above 800°C for 2hrs. μ-AES observation of the F added film with Ta = 920°C showed that some observation points had different compositions from the bulk one as shown in Fig. 2. Part 1 in the figure gave almost the same spectrum as the bulk one. The part 2 was Ba rich region and showed a presence of C probably due to $BaCO_3$. The part 3 was Cu rich region.

Superconducting Tc(onset) was in a range between 80 and 83 K as in the value of the film without

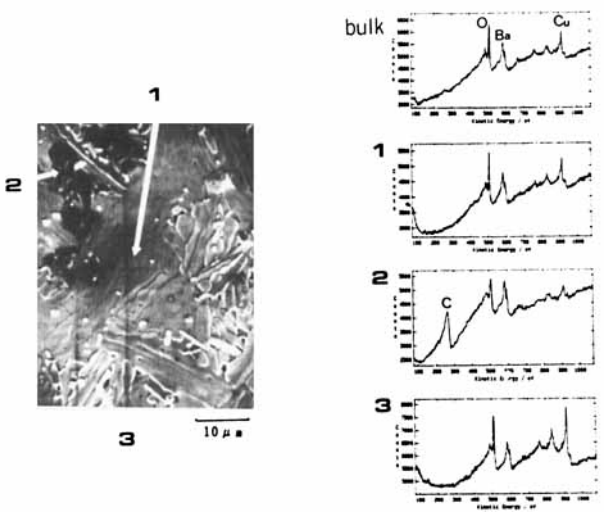

Fig. 2
SEM and micro auger electron spectrums of F added $YBa_2Cu_3O_y$ film /MgO.

fluoride. However its Tc(zero) was low around 60K probably because of a slight reaction with substrate. In order to keep fluoride in the film after heat treatment, the amorphous film/MgO was sealed in quartz ampoule with anhydrous FeF_3 and heated in a temperature range of 700°C to 1050°C. XRD show that the product contaminated with MgF_2 and that $YBa_2Cu_3O_y$ did not crystallize. Crystalline $YBa_2Cu_3O_y$ was observed on the heat-treated samples after a further treatment in oxygen flow at 920°C for 2hrs. YBCO crystallizes, therefore, as the crystals without fluoride in their composition.

Crystalline orientation on $SrTiO_3$ substrate was strongly affected by epitaxy. $YBa_2Cu_3O_y$ preferentially oriented its ‹100› along $SrTiO_3$ ‹100›, and its ‹110› along $SrTiO_3$ ‹110› in crystallization at 900°C of the amorphous film without F. Unknown product was observed at an interface between the substrate and the film annealed above 900°C. The substrate temperature was affected by applied rf power and target composition as shown in Table 1. The films as deposited were all in

X-ray amorphous state. They were annealed in oxygen flow in a temperature range of 880 ~ 900°C. YBCO crystal orientation changed with the deposition conditions of the amorphous film. The film deposited below Ts=260°C did not show any crystal orientation after its crystallization. Epitaxial crystalline orientation of YBCO crystal with $SrTiO_3$ substrate was observed in the film deposited from the normal target at rf power = 200W and also the film deposited from the fluoride added target at rf power = 100W after their annealing. Preferred orientation of YBCO (001) plane parallel to substrates was observed on the film deposited from the target with F at rf power = 200W. It is interesting that $YBa_2Cu_3O_y$ crystal orientation depends on the deposition condition of the amorphous film.

rf power	Normal target	Fluoride added target
100 W	Ts=260℃	Ts=300℃
	no orientation	YBCO<100>/$SrTiO_3$<100>
150 W		Ts=360℃
		YBCO<001>,<100>/$SrTiO_3$<100>
200 W	Ts=360℃	Ts=420℃
	YBCO<100>/$SrTiO_3$<100>	YBCO<001>/$SrTiO_3$<100>

Table 1. Preferred orientations of YBCO thin film/$SrTiO_3$(100) in relations to kinds of target and substrate temperatures Ts heated by rf sputtering (Ta=880 ~ 900°C).

REFERENCES

1. K. Nobugai and F. Kanamaru, Nihon-kagaku-kaishi, 2242 (1987).
2. F. Kanamaru, K. Oda, T. Yoshio and K. Takahashi, Mat. Res. Bull., 15, 525 (1980).
3. S. R. Ovshinsky, R. T. Young, D. D. Allred, G. DeMaggio and G. A. Van der Leeden, Phy. Rev. Lett., 58, 2579 (1987).
4. N. Moriyama, S. Kikkawa, M. Takahashi, K. Nobugai and F. Kanamaru, J. Jpn. Soc. Powder & Powder Metallurgy, 36, 579 (1989).