Pulsed-laser beam profiling for deposition of high-\( T_c \) YBCO films

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Abstract

A special shaping of YAG:Nd\(^{3+}\) laser beam profile has been found to improve homogeneity and \( T_c \) of YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) films being deposited by the pulsed laser. Targets of a considerably non-stoichiometric composition and substrates of slanting cuts have turned to be necessary for the high-quality film growing. Multiple droplets and solid particles ejected from the target surface by the infrared radiation are removed successfully from the laser-induced plasma by use of a velocity filter.

Key words: Films; laser deposition; high-\( T_c \) superconductor

1. Introduction

Conventional on-axis (as opposed to off-axis [1]) pulsed laser deposition is one of the most convenient methods for growing films of high-temperature superconductors (HTS) [2,3] and HTS-based multilayer structures [4].

A large number of droplets and other macro-particles arriving at the substrates is the main problem of the method. Use of a fast shutter for velocity filtration of plasma streams induced by a short-wave excimer laser radiation (wavelength 308 nm or shorter) has been found to be an effective method for protecting substrates from hit of the particles [5].

As compared to the excimer lasers YAG:Nd\(^{3+}\) lasers are handy and cheaper in use however the radiation density across their beams usually is non-uniform. Furthermore, they produce radiation of longer (1.06 µm) wavelength and generate much more droplets and solid particles of larger size, therefore.

In this paper new conditions of on-axis preparation of YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) (YBCO) films by YAG:Nd\(^{3+}\) pulsed laser are studied.

2. Experiment

The YBCO film deposition was performed in oxygen atmosphere at a pressure of 0.3 mbar on ZrO\(_2\):Y\(_2\)O\(_3\) (YSZ) and SrTiO\(_3\) single-crystal substrates heated to a temperature of 680\(^{\circ}\)C and placed at a distance 5.5 cm from a rotated Y-Ba-Cu-O ceramic target. A disc-chopper installed between the target and the substrate at a distance 3.8 cm from the target was used as a fast shutter. An opening of 2.5 cm diameter was milled out 6 cm off the disk center. The YAG:Nd\(^{3+}\) laser pulses of 10 ns duration, 0.1-0.4 J energy,
and about 14 Hz repetition rate were triggered by a phase-adjustable electronic device when the opening situated opposite the substrate. \( T_c \) of the films was measured as the temperature of the alternating-magnetic-field half-screening (i.e. when the susceptibility \( \chi' = -0.5 \)).

3. Results

Starting the on-axis deposition with targets of \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) composition we got non-stoichiometric films with low \( T_c \) and a large amount of precipitates on the surfaces. Changing the target composition by small steps we tested the properties of the films deposited in various conditions. The highest \( T_c \) and no precipitates had the films of nearly stoichiometric (or with a small Ba deficit) composition, however we found that their preparation required the targets of about \( \text{YBa}_{1.5}\text{Cu}_2\text{O}_x \) composition.

The ordinary on-axis deposition of 200 nm thick films using the infrared exciting radiation led to extremely large number of droplets (up to \( 10^8 \text{ cm}^{-2} \)). As the size of the particles was typically of 1 \( \mu \text{m} \) and reached 5-10 \( \mu \text{m} \), they covered the film surface almost completely and caused a dramatic \( T_c \) reduction. The disc-chopper rotated with a revolution rate higher than 200 Hz demonstrated an excellent separation of the fast vapor stream and the slow macro-particles by time of flight, the \( T_c \) increasing (see Fig. 1) and the surface becoming smooth. Practically complete detaining the particles (reduction by a factor of \( 10^6 \), at least) was observed with the rotation faster than 300 Hz.

We revealed also a dramatic \( T_c \) reduction with increasing non-uniformity of the radiation density. So far as highly uniform beam profile was difficult to achieve and maintain, we increased the mean pulse energy density from 5-8 to 10-20 J/cm\(^2\) so reducing the vapor composition fluctuations due to complete evaporation of the less-irradiated target zones. However, it led to a ball-like plasma shape and to bad transfer of the material to the substrate.

![Fig. 1. \( T_c \) of the YBCO films grown on YSZ substrates with different rotation frequency \( n \) of the disc-chopper.](image1)

By various multimode adjustment of the laser resonator or using a mask we formed the beam profile so that to get sharp and bright lines (\( \bigtriangledown, \times, \bigcirc \)) or several bright spots instead of the ordinary (\( \bullet \)) filled-circle shape. As a result we got both the high pulse energy density and the large size of the irradiated target area without increasing the entire energy, so providing an oblong shape of the plasma and an effective vapor transfer. A reproducible growth of the high-quality films was enabled (see examples in Table 1 and Fig. 2).

![Table 1](image2)

**Table 1**

| Beam profile | \( \bullet \) | \( \bullet \) | \( \bigtriangledown \) | \( \times \) | \( \bigcirc \) |
|--------------|-------------|-------------|-------------|-------------|-------------|
| Substrate    | YSZ         | SrTiO\(_3\) | YSZ         | SrTiO\(_3\) | SrTiO\(_3\) |
| \( T_c, K \) | 77.0        | 81.4        | 90.1        | 91.3        | 91.0        |
|              | 86.1        | 88.0        | 90.2        | 90.8        | 91.4        |
|              | 82.7        | 89.2        | 89.9        | 91.0        | 90.3        |
|              | 87.1        | 77.5        | 90.0        | 90.7        | 90.2        |

![Fig. 2. Alternating-field screening curves of YBCO films grown on YSZ (\( \bigtriangledown \)-profiled beam, \( n = 400 \text{ Hz} \)) and SrTiO\(_3\) (\( \times \)-profiling, \( n = 380 \text{ Hz} \)) substrates.](image3)

Since our films grown on highly \( (100) \)-oriented substrates had reduced \( T_c \) (85-87 K), to all appear-
ance due to the terrace epitaxy disturbance [6], we became using slanting cut substrates (all the data presented above belong to the films grown on the substrates cut 2-4° off the (100) plane). Stable growth of YBCO films with $T_c$ about 90 K or higher was observed on YSZ and SrTiO$_3$ substrates cut with the slant from 2 to 20°.

4. Conclusion

In summary, due to the foregoing study of the on-axis YAG:Nd$^{3+}$ laser deposition of HTS films, several novelties (namely, the beam profiling, the velocity filtering realized for the infrared exiting radiation, the considerably non-stoichiometric targets, and the slanting cut substrates) have been introduced and reproducible growth of high-quality YBCO films has been reached.

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References

[1] B. Holzapfel, B. Roas, L. Schultz, P. Bauer, and G. Saemann-Ischenko, Off-axis laser deposition of YBa$_2$Cu$_3$O$_{7-\delta}$ thin films. Appl. Phys. Lett. 61, 3178-80 (1992).
[2] B. Roas, L. Schultz, and G. Endres, Epitaxial growth of YBa$_2$Cu$_3$O$_{7-x}$ thin films by a laser evaporation process. Appl. Phys. Lett. 53, 1557-59 (1988).
[3] A. I. Golovashkin, E. V. Ekinov, S. I. Krasnosvobodtsev, E. V. Pechen, Single-crystal Y(Eu, Ho)Ba$_2$Cu$_3$O$_7$ films. Physica C 153-155, 1455-56 (1988).
[4] E. V. Pechen, R. Schönberger, B. Brunner, S. Ritzinger, K. F. Renk, M. V. Sidorov, and S. R. Oktyabrsky, Epitaxial growth of YBa$_2$Cu$_3$O$_{7-\delta}$ films on oxidized silicon with yttria- and zirconia-based buffer layers. J. Appl. Phys. 74, 3614-16 (1993).
[5] E. V. Pechen, A. V. Varlashkin, S. I. Krasnosvobodtsev, B. Brunner, and K. F. Renk, Pulsed-laser deposition of smooth high-$T_c$ superconducting films using a synchronous velocity filter. Appl. Phys. Lett. 66, 2292-94 (1995).
[6] R. Grosser, A. Martin, O. Kus, E. V. Pechen, and W. Schöpe, Levitation of small permanent magnet between superconducting YBa$_2$Cu$_3$O$_{7-\delta}$ thin films. Proc. Appl. Supercond. Int. Conf. (The Netherlands, 30 June-3 July 1997), Inst. Phys. Conf. Ser. 158, 1643-46 (1997).