

Thermally-induced self-assembling nanotechnology of gold nano-dots on CeO₂-buffered sapphire for superconducting films

A. CRISAN^{a,b,*}, R. WÖRDENWEBER^c, E. HOLLMANN^c, R. KUTZNER^c, T. W. BUTTON^a, J. S. ABELL^a

^aDepartment of Metallurgy and Materials, University of Birmingham, Birmingham B15 2TT, United Kingdom

^bNational Institute of Materials Physics, Bucharest, 077125 Romania

^cInstitute of Thin Films and Interfaces, Research Centre Jülich, Germany

A typical facility for fabrication of devices based on high temperature superconducting (HTS) films grown on the less-expensive CeO₂-buffered sapphire substrates comprises a transfer chamber, a chamber for CeO₂ growth, a chamber for YBa₂Cu₃O_y (YBCO) growth, and a chamber for Au deposition at room temperature. Here we present the adaptation of our previously-reported method to create artificial pinning centres in HTS films through self-assembled nano-scale islands for this type of facility without making any modifications. We succeeded to grow Au nano-dots from an ultra-thin layer of Au grown at room temperature subsequently annealed in the chamber for YBCO deposition, in the same conditions as for YBCO growth. AFM and RBS were used to investigate the formation of Au nano-dots, while XRD and susceptibility measurements were used to study the properties of nano-structured YBCO films.

(Received April 23, 2008; accepted June 4, 2008)

Keywords: Applied superconductivity, YBCO thin films, Self-assembling nanotechnology, Au nano-dots.

1. Introduction

Not long after the discovery of high-temperature superconductors (HTS), a world-wide research effort led to the demonstration of a wide range of applications, based both on HTS thin films (cryoelectronic devices) and on HTS wires, tapes and conductors (power devices).

The most important factor limiting the performance (low-frequency noise and sensitivity in active devices, quality factor and power handling capabilities in passive microwave components, critical current density J_c in power applications) is the thermally-activated movement of magnetic flux lines (vortices), that can be suppressed by sufficiently strong pinning centres in the HTS material. So, engineering of artificial pinning centres and fundamental studies regarding their effect on the vortex dynamics and other superconducting (SC) properties are of great importance for science and technology of HTS.

Recently, it was proposed and proved [1-4] a cost-effective and straightforward method for creating strong pinning centres in HTS thin films by self-assembled growth of three-dimensional (3D) nano-scale dots on substrates, prior to the film deposition. Preliminary results showed the huge potential of the new technology for improving the SC properties of thin films (increased J_c 10-20 times in a wide range of applied magnetic field, increased pinning potential U_p several times and decreased rate of thermally-activated flux creep by two or more orders of magnitude). For cryoelectronics applications of HTS films (including microwave devices), the most commonly used substrate is sapphire with a thin CeO₂ buffer layer.

The purpose of this work is to adapt the above-mentioned technology for the case of a typical fabrication facility comprised of a transfer chamber, a chamber for CeO₂ growth, a chamber for YBa₂Cu₃O_y (YBCO) growth,

and a chamber for Au deposition at room temperature for electrical contacts, *without making any modifications*. In this respect, the main goal was to grow Au nano-dots from an ultra-thin layer of Au grown at room temperature subsequently annealed in the chamber for YBCO deposition, *in the same conditions of temperature and atmosphere as for YBCO growth*.

CeO₂ buffer layers, Au ultra-thin layers and nano-dots were characterized by Atomic Force Microscopy (AFM) and Rutherford Back-Scattering (RBS), while the structural and superconducting properties of YBCO HTS films were studied by X-ray diffraction (XRD) and susceptibility measurements.

2. Experimental

CeO₂ buffer layers (about 30-50 nm thick) were grown on r-cut sapphire (3-4'' in diameter) by rf magnetron sputtering in the dedicated chamber. Then the substrates were cut into 1 cm squares, cleaned, and used for further steps. Ultra-thin layers of Au were grown at room temperature by dc sputtering in the dedicated chamber, at several low-values of discharge power (10, 20, 50 W) and with several deposition times (5-10 sec) on sapphire/CeO₂ substrates. Several substrates were used for each deposition, and on each substrate a mask was used to avoid Au deposition on *half of it, for comparison*. From each batch, some substrates were used for AFM and RBS, some were annealed and studied by AFM and RBS, and on some substrates (with annealed Au), YBCO HTS films were grown by rf sputtering. Gold annealing was performed in the chamber for YBCO deposition, in the same conditions (temperature, atmosphere) as for YBCO growth: three temperature steps at 250 °C, 650 °C, and 843 °C, with the total time for temperature increase of

about 1 hour, and the same Ar/O₂ mixture and pressure as for YBCO growth. YBCO films grown in the same conditions on substrates with Au nano-dots of various architecture and on substrates without nano-dots were characterized by XRD and low-field ac susceptibility.

3. Results and discussion

From the first RBS studies we were able to correlate the dc power and deposition time with the resulting thickness of Au ultra-thin layers, and select the deposition parameters suitable for the subsequent fabrication of Au nano-islands. With increasing dc power and/or deposition time, in RBS spectra the height of the Au-peak (smaller peak at about 129 MeV) increases (Au layers become thicker), while the CeO₂ peak (larger peak at about 124 MeV) shifts to lower energies (incident particles are losing more energy in the Au layer before reaching the CeO₂ layer), as can be seen in Figure 1(a).

For small dc power and deposition time of Au, the thermal treatment in the same conditions as those for YBCO growth led to the formation of Au nano-dots on top of the CeO₂ buffer layer. The comparison of RBS spectra of two identical sapphire/CeO₂/Au samples, one as grown and the second one annealed is shown in the Fig. 1(b).

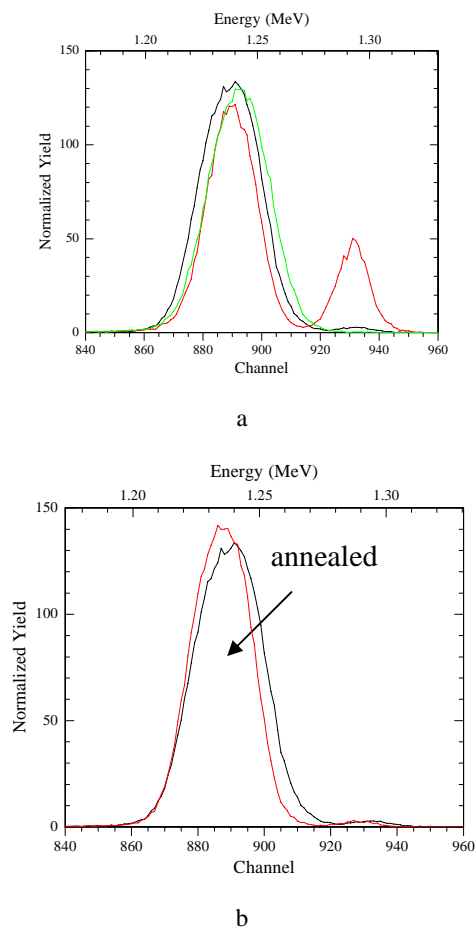


Fig. 1. High-energy RBS spectra of sapphire/CeO₂/Au samples: (a) different Au thickness, and (b) same (ultra-thin) Au thickness, as-grown and annealed.

The results shown in Fig. 1(b) are quite important: due to annealing, the CeO₂ peak moves back towards higher energy (almost in the same position as in the case of substrates without Au), meaning that, after annealing, most of the CeO₂ surface becomes un-covered by Au, and incident particles reach CeO₂ without loss of energy. This is an indirect evidence of Au nano-dots self-assembling through thermal annealing from the initial ultra-thin (nominally 2-3 unit cells) Au film.

Direct evidence of this process is clearly provided by AFM. Figure 2 shows some examples of AFM images, taken on a sample with as-grown Au and on a sample with the same thickness of Au, annealed, on various locations.

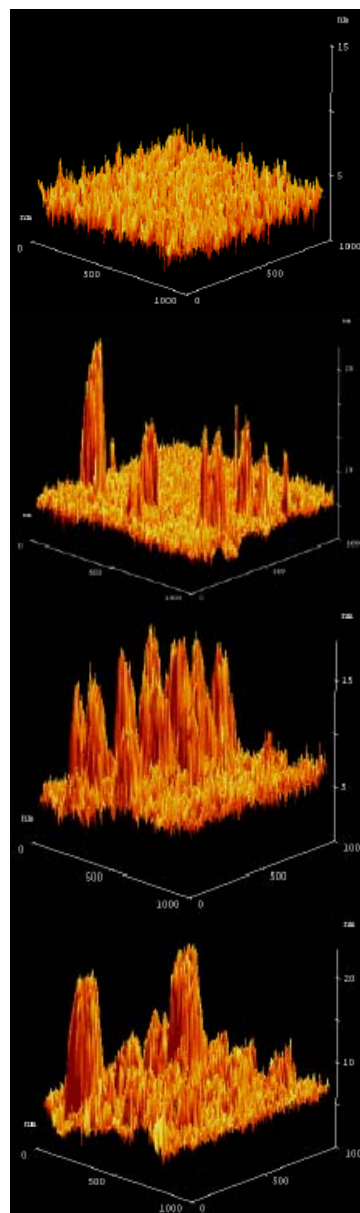
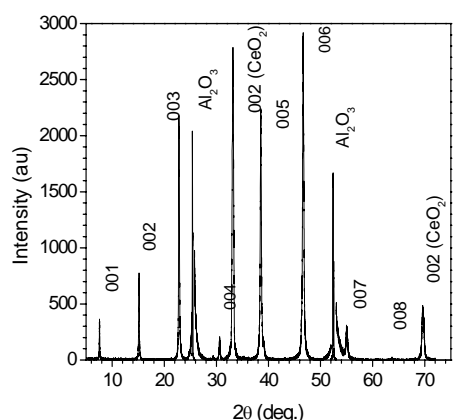


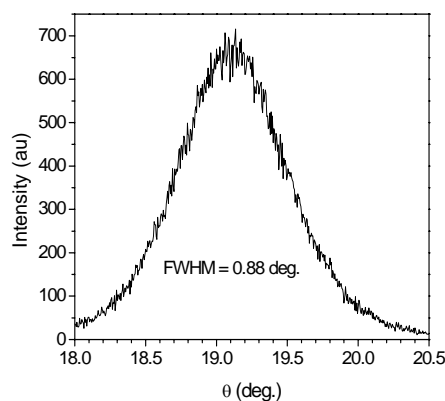
Fig. 2. AFM images of ultra-thin Au (top), and of Au nano-dots, on various locations on the same sample.

As can be seen, Au nano-dots are randomly distributed, with relatively random density, with in-plane diameters between 40 and 100 nm, and height between 10 and 20 nm.

The crystalline quality of the YBCO films grown on substrates with various architectures of Au nano-dots, and, for comparison, on substrates without Au was studied by XRD, namely Bragg-Brentano (θ - 2θ) scans and ϕ scans of the (005) reflection. There was no notably difference between YBCO films grown on substrates with Au nano-dots and films grown on bare substrates. In Figure 3 is shown an example of XRD patterns, for a YBCO film grown on sapphire/CeO₂/Au substrate similar to the one shown in Fig. 2.



a



b

Fig. 3. XRD patterns of Al₂O₃/CeO₂/Au nano-dots/YBCO: θ - 2θ scan (top), and rocking curve (ϕ scan) of the YBCO film (005) reflection (bottom).

It can be clearly seen the excellent epitaxial growth (only 00l reflections) of the YBCO, as well as the quite low (considering the cheap sapphire substrate) mosaic spread (FWHM of about 0.88 deg.). Note that the curious

shape and low intensity of the Al₂O₃ substrate reflections is due to an automatic decrease of detector's gain performed by the diffractometer at very high amplitudes of the signal (without this, the signal from the substrate would have been much higher).

The superconducting properties of YBCO films grown on substrates with various architectures of Au nano-dots, and, for comparison, on substrates without Au were investigated by ac susceptibility measurements with low ac fields amplitude. Temperature-dependence of the in-phase ac susceptibility response of the investigated YBCO films is shown in Fig. 4.

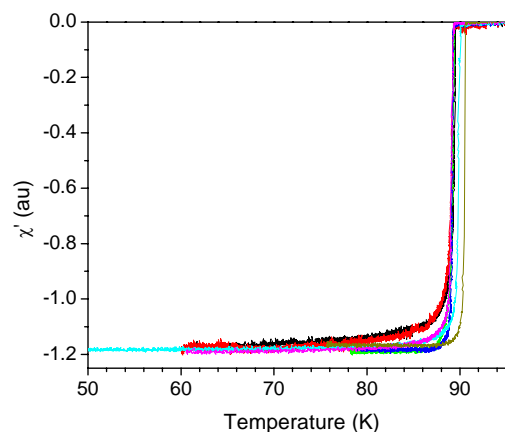


Fig. 4. Temperature dependence of in-phase ac susceptibility response of several YBCO films, with and without Au nano-dots.

It can be seen that all films have sharp superconducting transitions with the critical temperatures T_c of 90 ± 0.5 K, depending on increasing or decreasing temperature of measurements. The influence of Au nano-dots density and dimensions on other superconducting properties like critical current density and pinning potential is currently under investigation.

4. Conclusions

In conclusion, we have shown that the previously-reported self-assembling nanotechnology of pinning centres in superconducting thin films can be easily adapted, without any modification, to typical facilities of cryoelectronic devices fabrication. We have shown that Au nano-dots can be fabricated from ultra-thin layers of Au by thermal annealing in the same conditions like those needed for YBCO growth. The crystal quality, critical temperature, and sharpness of superconducting transition of YBCO film were not influenced by Au nano-dots. This technology can be applied, depending on the influence of nano-dots architecture on critical current density, either for

increasing the quality of cryoelectronic devices, or for the fabrication of magnetic flux ratchet devices.

Acknowledgment

This work was supported by the European Science Foundation (“THIOX” Programme), by the European Commission through the Marie Curie Excellence Grant “NanoTechPinningHTS”, and by the Romanian Ministry of Education and Research.

References

- [1] A. Crisan, S. Fujiwara, J. C. Nie, A. Sundaresan, H. Ihara - Appl. Phys. Lett. **79**, 4547 (2001).
- [2] A. Crisan, P. Badica, S. Fujiwara, J.C. Nie, A. Sundaresan, A. Iyo, Y. Tanaka - IEEE Trans. Appl. Supercond. **13**, 3726 (2003).
- [3] M. Ionescu, A. H. Li, Y. Zhao, H. K. Liu, A. Crisan J. Phys. D: Appl. Phys. **37**, 1824 (2004).
- [4] A. Crisan, P. Badica, S. Fujiwara, J. C. Nie, A. Sundaresan, Y. Tanaka, H. Ihara, Appl. Phys. Lett. **80**, 3566 (2002).

*Corresponding author: i.a.crisan@bham.ac.uk