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Raman Spectroscopic Studies of $YBa_2Cu_3O_7$ Coated Conductors

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$YBa_2Cu_3O_7$ Coated Conductors의 Raman 분광학 연구

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Abstract

We present results of Raman spectroscopic studies of superconducting $YBa_2Cu_3O_7$ (YBCO) coated conductors. Raman scattering is used to characterize optical phonon modes, oxygen content, c-axis misalignment, and second phases of the YBCO coated conductors at a micro scale. A two-dimensional mapping of Raman spectra with transport properties has been performed to elucidate the effect of local propertied on current path and superconducting phase. The information taken from the local measurement will be useful for optimizing the process condition.

Keywords : Raman spectroscopy, YBCO coated conductors, critical current density

I. Introduction

Various processing methods are being investigated for the fabrication of high-quality current density (J_c) YBCO coated conductors on metal tape substrates. Vacuum processes such as pulsed laser deposition (PLD) [1,2], e-beam co-evaporation [3], and metal-organic chemical vapor deposition [4] are generally producing high-J_c films. Usually, the associated seed/buffer layers are also prepared by vacuum processes such as ion-beam assisted deposition (IBAD) [5].

When considering long-length continuous process for conductor production, highly-controllable vacuum processes are promising. When considering long-length continuous process for conductor production, highly-controllable vacuum processes are promising. In particular, PLD is demonstrated to be a front-line technique which has accomplished the best results so far in terms of critical current density in a long-length conductor.

One of the important requirements for coated conductor applications is to maximize the current carrying capacity in a given width of conductor. Vexingly, the common experience is that the total current carrying capacity of YBCO coated conductors does not scale linearly with the thickness and the length of the conductor, quite independent of the particular process used to deposit the conductor.

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Correspondingly, the average critical current density of such films decreases as the thickness and the length of the film increases. The reason for this behavior is being studied by several groups [6-8] by not yet understood. The thickness and the length-dependence could result from variations in the material properties as a function of thickness and length, or it could result from intrinsic properties of collective pinning, or both.

In spite of the importance of YBCO for application technology, suitable tools for accurate absolute evaluation of YBCO films are not enough and lacking. Raman spectroscopy has proved to be a valuable tool in investigations of the physical properties of high-T_c superconductors [9-11]. Most studies have concerned the frequencies, line-shapes and intensities of $k \approx 0$ phonons, and how these phonon parameters depend on structure, chemical composition, temperature, pressure and the interaction with other elementary excitations. Raman scattering spectroscopy has also been used as a complementary method for chemical and structural characterization of high-T_c superconductors [9,11]. In this paper, we address our careful attention to the various useful capabilities of the confocal micro-Raman scattering technique for quality characterization of YBCO coated conductors. In particular, we have an interest in obtaining local information on optical phonon mode, oxygen content, misalignment grains, and second phase formation, which might be essential for determining critical current density.

II. Experiment

YBCO films were prepared on CeO_2/ IBAD-YSZ coated metal tapes by a reel-to-reel pulsed laser deposition (PLD) method. It is shown by x-ray diffraction that the as-grown YBCO films have a highly c-axis oriented and in-plane aligned texture. The critical current densities and the thickness of the YBCO layers are in the range of 0.2 ~ 1 MA/cm^2 and 0.7 ~ 1 MA/cm^2, respectively.

Samples were thermally treated by a double or triple annealing process. During the deposition process, the substrate temperature of sample #1 and #2 was set in 807°C for the bottom half layer. For growing of the top half layer, #1 and #2 samples were experienced temperature 835 °C and 870 °C, respectively. In case of the samples #3 and #4 one third of the films were grown at 807 °C and central parts of the films were grown at 840 °C. After all, the samples #3 and #4 were grown at 840 °C and 880 °C, respectively. The reason we increased temperature for the second and third deposition is that higher growth temperature is required to get c-axis oriented texture for the multi-coating process.

Raman scattering measurement was performed in the backscattering geometry using a triple mode Raman spectrometer (Jobin Yvon T64000). The optical arrangements for all Raman measurements were in the backscattering geometries with the incident light propagating normal to the film plane. In the scattering experiment, the excitation direction of polarization was X and scattered light was detected without a polarizer. Skin depth of optical beam into YBCO layers is a few hundreds of nanometer, depending on resistivity of the YBCO layers. The incident laser beam was the 514.5nm line of an Ar-ion laser. The sampling area was focused by the laser beam with a magnification of 50. The laser power was low enough to prevent sample heating.

We could measure optical properties of YBCO films over tens of micrometer on the surface. Spectral resolution of the experiment was 1.2cm^{-1}. All of measurements were carried out at room temperature. In general it took 90 minutes (3 cycles) to measure the optical spectra of YBCO films.

III. Results and Discussion

Figure 1 presents the Raman spectra of YBCO coated conductors. In this case, four samples were used by single mode. They all show typical 3 YBCO phonon modes at the ~340 cm^{-1}(B1g), ~440 cm^{-1}(A1g), ~500cm^{-1}(AO), which are attributed to out-of-phase bending bond on the CuO_2 plane, in-phase bending bond on the CuO_2 plane and stretching bond of O-O in the apical. Through exhibiting 500cm^{-1} at all mode, we could insist that each sample has the different amount of oxygen.

The peak frequency of the AO band provides an

approximate measure of the oxygen stoichiometry of YBCO. For tetragonal $YBa_2Cu_3O_x$, x is expected to be near a value of 6 and the AO peak frequency is near 480cm^{-1}, whereas for the orthorhombic form x is expected to be near 7.0 and the AO mode peak frequency is near 500cm^{-1} for all of the samples, indicating that the YBCO phase is the orthorhombic form in all cases.

Now it needs to concern about some extraordinary peaks. The peak of the 627cm^{-1}(BC) bands with sample #1, #2 and #3 is related to $BaCuO_2$[9,11]. $BaCuO_2$ phase has broad band especially at sample #3. In case of ~600cm^{-1} mode, it could be attributed to $Ba_2Cu_3O_{5.9}$, indicating that the $Ba_2Cu_3O_{5.9}$ had changed into oxygen-lack $BaCuO_2$[9].

The band marked CM shown in Fig. 1 is the O4 phonon of YBCO, suggesting that there is non c-axis oriented grains in the film. In the xx/yy configuration this mode should have minimal intensity if the YBCO film is perfectly epitaxial. The extent to which this mode appears relative to the 340cm^{-1} phonon is in essence a measure of the degree of c-axis misalignment. Using this relationship as a guide, it is found that that c-axis misalignment is displayed in the #4 sample. In Table 1, a summary of Raman peak intensities is displayed with transport information of the films. It is plausible to extract correlation between high critical current density and 500cm^{-1} mode etc. Also, existence of peaks larger than 600cm^{-1} could be linked to low critical current density. A more quantitative analysis is under way.

Table 1. Information of the samples used in the Raman scattering measurements and the peak intensities of Raman spectra. Film thickness of sample #1, #2, #3 and #4 is 700nm, 700nm, 1050nm and 1050nm respectively.

Sample ID	Raman Frequency (cm^{-1})					Ic (A/cm)	Jc (x10^{6}A/cm^2)
	335	435	500	594	627		
#1	518	374	1130	231	131	69.5	0.99
#2	5240	1860	2730	1480	1200	18.5	0.26
#3	3460	720	2030	321	742	37.5	0.35
#4	4430	790	742	3910	792	17.5	0.17

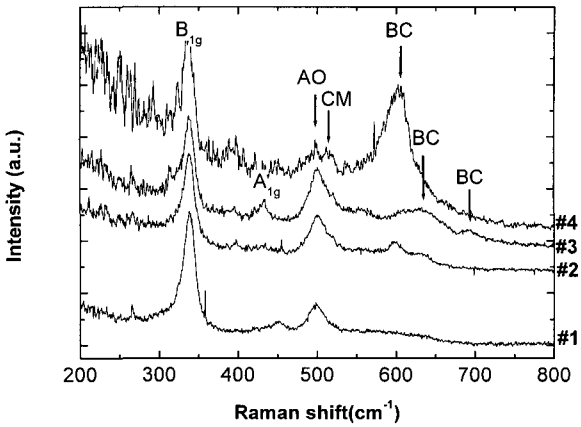

Fig. 1. Raman spectrum of YBCO films prepared by different method: B_{1g}: YBCO Cu-O at plane; A_{1g}: YBCO Cu-O at plane; AO: apical oxygen; CM: YBCO c-axis misalignment indicator mode; BC: Ba-Cu-O phonon.

In Fig. 2, two-dimensional mapping of Raman spectra has been performed with sample #3. The information from Fig. 1 was taken over an arbitrary position, indicating that the spectra are not considered local variations. Interpretation of each Raman peak is vital for judging good or bad part of YBCO films relating to current path and superconducting phase. X designates a position of width of the conductor, and Y a location of length of the conductor. So, x = 1, 3 mm corresponds to regions close to edge whereas x = 2 mm means that central part of the conductor. As shown in Fig. 2, it is quite surprising that the spectra show a very much different behavior over the whole regions of the conductor. In particular the peak intensities near 500 and 650 cm^{-1} are a lot different, suggesting that the local transport properties of the regions are different. In other words, the region with 500 cm^{-1} will carry large current and the region with 650 cm^{-1} will carry almost no current. The region mixed with both the peaks will show intermediate behavior. A local transport measurement is under progress to verify this statement.

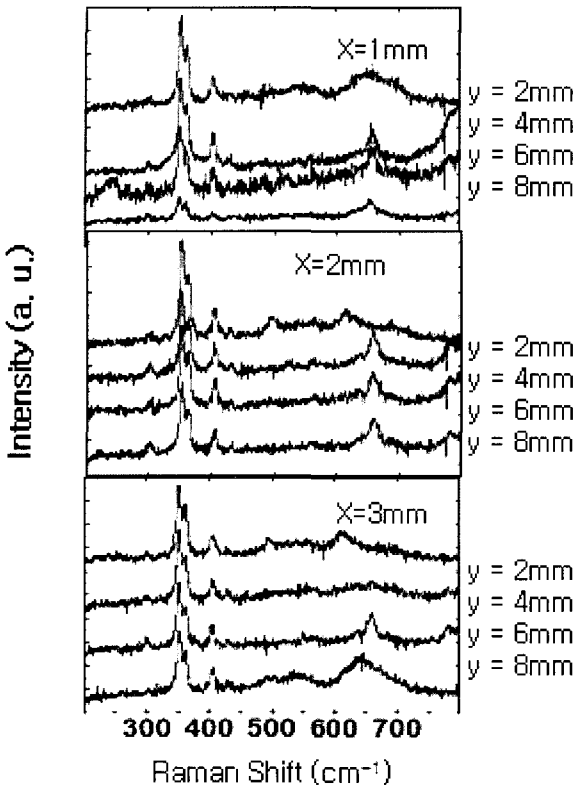

Fig. 2. Two-dimensional mapping of Raman spectra of sample #3. Dimension of the YBCO film is 4mm, 10mm in direction of x and y, respectively.

IV. Conclusion

YBCO coated conductors prepared by a reel-to-reel pulsed laser deposition are measured through confocal micro-Raman spectroscopy. Extra peaks around 500, 600, 650 cm^{-1} including YBCO phonon modes reveal that oxygen content, c-axis misalignment, and second phases of the YBCO coated conductor. Two-dimensional Raman mapping method was demonstrated in order to find out its optimal processing conditions and local properties in micron scale. Through quite different spectra, we could rationalize the effect of local properties on current path and superconducting phase. We could also discover that in even the same sample quite different phases coexist. From the results, it is expected to know the local properties of each site in the same sample in detail to get optimistic processing conditions.

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