

PULSED LASER DEPOSITION OF HIGH T_c SUPERCONDUCTING THIN FILMS: PRESENT AND FUTURE

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ABSTRACT

Pulsed laser deposition (PLD) has been widely used for deposition of high T_c superconducting thin films, and has been approved as one of the best physical vapor techniques to prepare the films. The most important advantage of this technique is stoichiometric deposition, namely the films could be made with the same composition of the targets. Utilized PLD, not only thin films but also multilayers and superlattices of high T_c superconductors have been fabricated. In this paper, the performance of the technique will be reviewed, and the future will be speculated.

INTRODUCTION

Utilizing laser irradiation as an energy source to vaporize materials and form thin films has been researched over the last twenty years. An excellent review has been given by Cheung and Sankur in 1988 [1]. The successful use of laser deposition of high T_c superconducting thin films has brought world-wide attention to this less well known thin film deposition technique. Laser deposition is becoming a strong contender among various vapor deposition techniques for deposition of not only superconducting thin films, insulators, and metals but also for multilayers and superlattices.

CHOICE OF LASERS

There are various lasers (both pulsed and cw) with wavelengths from ultraviolet to far-infrared are commonly available. The choice of the lasers depends on specific applications as discussed by Boyd [2]. In applications where melting and evaporation are desired, the pulsed lasers have been selected because of their high power outputs.

The absorption coefficient of the material plays an important role in the laser-material interaction process [1-2]. The higher the absorption coefficient, the thinner surface layer will be heated to the vaporization point. Moreover, the reflectivity should be small in order to incorporate the optical energy to the material more efficiently. In table 1, estimated values of absorption coefficient and reflectivity for $YBa_2Cu_3O_{7-x}$ are given for few selected wavelengths. Those values are calculated from the data of optic transmission or ellipsometry measurement on $YBa_2Cu_3O_{7-x}$ superconducting thin films [3-5]. The values could be different for bulk materials. The large absorption coefficient and small reflectivity at 193 nm, 248 nm and 308 nm make excimer lasers the choice for deposition of high T_c superconducting thin films. Though large wavelength lasers such as 1064 nm Yd:YAG laser were employed for thin film deposition, the superconducting properties of the deposited films were generally worse than those deposited using excimer lasers [5-6].

SUPERCONDUCTORS

Which high T_c superconductor should be the choice for target material? As we know, there are a number of Cu-based oxides which are superconducting over 77 K [7-9].

Table 1. Estimated values of absorption coefficient (α) and reflectivity (R) for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$.

Laser	Wavelength (μm)	α (10^5 cm^{-1})	R
Carbon Dioxide	~ 10	0.5	0.75
Nd:YAG	1.064	1.2	0.18
Nd:YAG ($\lambda/2$)	0.533	1.5	0.14
Nd:YAG ($\lambda/3$)	0.355	1.7	0.12
Excimer			
XeCl	0.308	1.9	0.12
KrF	0.248	2.3	0.13
ArF	0.193	2.4	0.15

From an application point of view, a higher superconducting transition temperature would be desired. So far, Tl-based superconductors have the highest transition temperature of 125 K [9]. The main problem for making Tl-base oxide superconducting thin film is not the toxic issue of the materials but the existence of many superconducting phases which are formed at similar processing conditions. This creates difficulties to form single phase superconducting thin films with higher transition temperatures. Bi-based superconductors have the same problem. Loss of Tl and Bi at high substrate temperatures also introduces extra problems for thin film fabrication. That is why so far there are no good superconducting thin films with transition temperatures over 100 K. Certainly, it is a challenge to solve the mentioned problems.

At this moment, the choice of superconductors for thin film fabrication is 90 K $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$ (Re=Y, Dy, Ho, Er, Gd, ...) (so called "123" superconductors). The "123" superconducting thin films are much easier to be formed in a single phase. Many groups are able to make "123" superconducting thin film with critical current densities of $5 \times 10^6 \text{ A/cm}^2$ at 77 K, which is the highest for any superconductors. For this reason, we will restrict our discussions only for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBaCuO).

YBaCuO has an orthorhombic structure with $a=0.382 \text{ nm}$, $b=0.389 \text{ nm}$ and $c=1.168 \text{ nm}$. For the material to be superconducting, one must be able to (1) make films with right composition, (2) get right crystalline phase, and (3) produce the right oxygen stoichiometry in the films.

STOICHIOMETRIC DEPOSITION

Thin film deposition process can be divided into three steps: vaporization of the source material, transporting the vaporized material to substrate, and film nucleation on the substrate surface. The ideal way for thin film fabrication is to use a stoichiometric target, and obtain films with the same composition of the target.

Congruent evaporation is the most unique feature of pulsed laser deposition [1]. For a multicomponent material, the pulse heating enables all components to be evaporate instantly. Though pulsed laser deposition had been used for preparation of thin film of BaPbBiO [10], which has T_c of 13 K, it was first reported by Dijkkamp *et. al.* [11] that a multi-element oxide could be deposited stoichiometrically by pulsed excimer laser

deposition. Fig. 1 shows a Rutherford backscattering (RBS) spectrum of a Y-Ba-Cu oxide film on a carbon substrate (solid line) made from a stoichiometric $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ target at room temperature in vacuum. It is obvious that the film had a composition very close to the target as evident from the simulation (dashed line) assuming Y:Ba:Cu:O=1:2:3:6. The stoichiometric deposition indicates congruent evaporation of the superconductor by pulsed laser heating, and unite or same sticking coefficients for all the elements. The small loss of oxygen in vacuum is obvious. The large amount of oxygen found in the film deposited in vacuum shows strong evidence for existence of oxides or sub-oxides in the evaporated material, which was confirmed by a mass spectroscopic study [12].

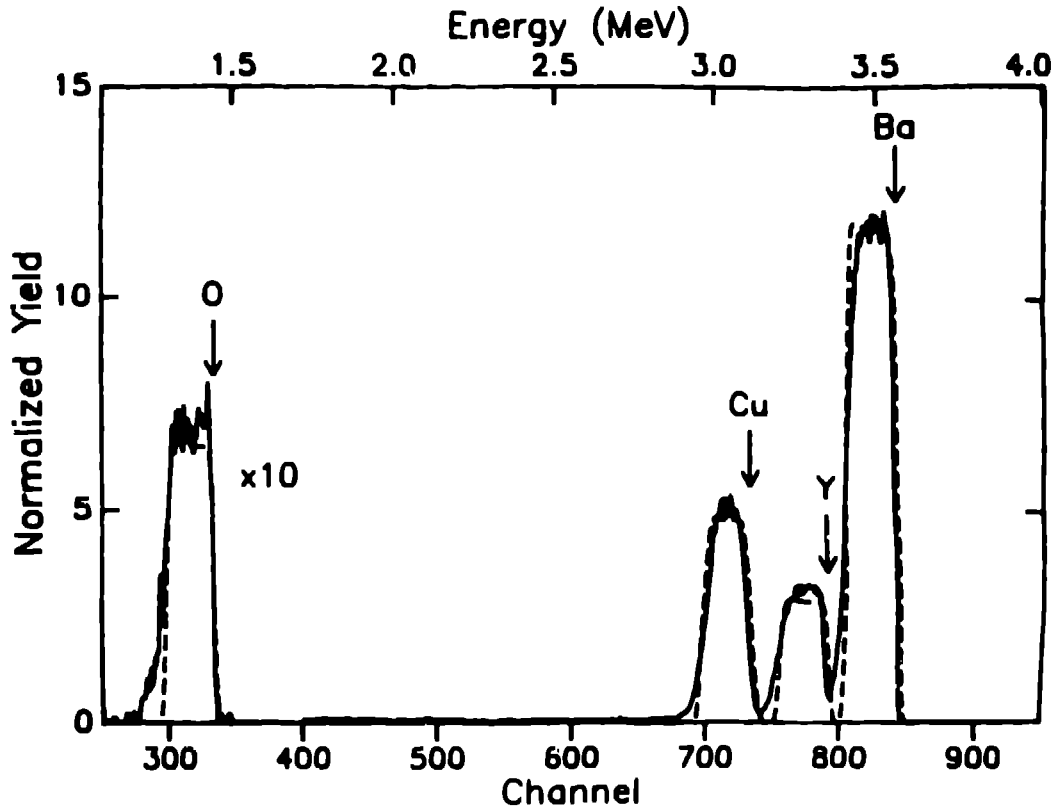


Fig. 1 RBS Spectrum (4 MeV He^{++}) of an as-deposited Y-Ba-Cu oxide thin film on a carbon substrate (solid line) made at room temperature in vacuum, overlaid with a simulation (dashed line) of 190 nm $\text{YBa}_2\text{Cu}_3\text{O}_6$.

The pulsed laser deposition process is rather complicated as shown by a further study. In the experiment [13], a pulsed laser (248 nm, 30 ns) was incident on a rotating target at an angle of 45 degrees. The substrate was held in parallel with the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ target. The angular distributions of composition and thickness of the deposited films were measured by RBS [13]. The data in Fig. 2 was obtained at an energy density of 1.5 J/cm^2 , and the angle was defined as from the normal direction of the target. The results clearly showed that the deposition was forward-directed, and the composition for large angles was deviated from the ideal one such as in the center of the deposition. It was also found that there is a laser energy density threshold ($\sim 1 \text{ J/cm}^2$ for the excimer laser) to obtain correct composition even at the center [13]. Since more particles (size up to 1000 nm in diameter, on the film surfaces) per unit thickness were found at higher energy densities, there is a window of laser energy density, typically $1.5\text{-}3 \text{ J/cm}^2$ for excimer lasers, for thin film deposition.

Furthermore, at fixed energy densities, the composition of the deposited films of $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$ (Re = Y, Dy, Ho, Er, Gd, ...) was not effected by the deposition chamber background pressure and the substrate temperature up to 800 C [14]. This unique feature of laser deposition makes thin film fabrication much easier.

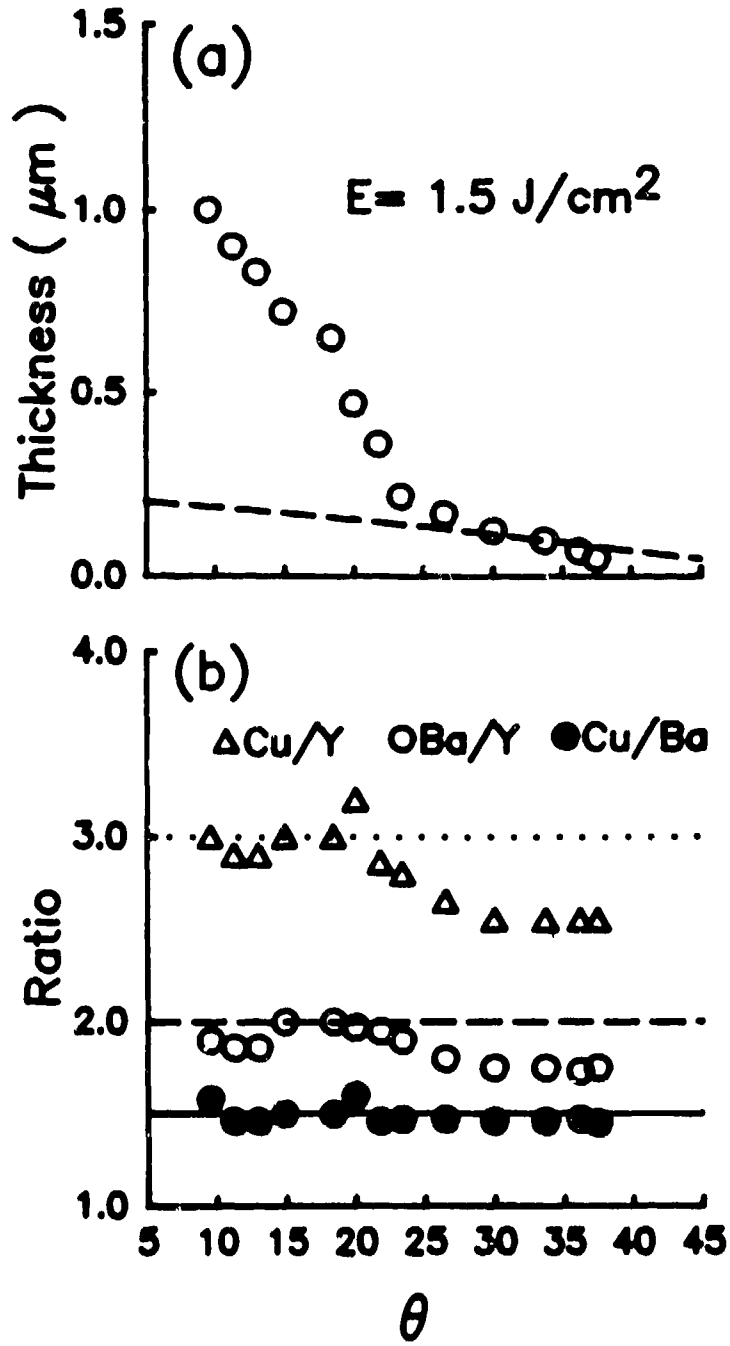


Fig. 2 At 1.5 J/cm^2 laser (248 nm, 30 ns) energy density, the angular distribution of the deposited film: (a) thickness (the dashed line is the $\cos\theta$ fit) and (b) composition.

SUPERCONDUCTING THIN FILMS

Once the correct composition in the films are obtained, there are still two obstacles to be overcome before one can have good superconducting films. The atoms in the film must be in the right crystallographic phase and the right amount of oxygen needs to be incorporated into the structure.

There are at least two approaches [3] to accomplishing this: one is to deposit the film in some random phases at a relatively low temperature and follow this up with a high temperature (800-900 C) anneal in oxygen. But the high temperature treatment creates a number of problems [15]:

1. chemical reaction with the substrate resulting in the formation of a dead layer at the interface,
2. loss of film stoichiometry after annealing due to film-substrate interaction, and
3. crack formation during thermal cycling destroying the percolation path for superconductivity in the film.

These lead to poor reproducibility and performance of the films.

A more elegant way to accomplish the goal is to deposit the films directly in the crystalline phase at a temperature of 600-700 C in an oxygen ambient. From a technological point of view, in order to produce sharp interfaces and minimize film-substrate interaction and stresses in the films, a low temperature process is absolutely essential. With the second approach epitaxial films with superior superconducting properties could be prepared since the films are grown layer-by-layer.

The first experiment on the low temperature process was done at a deposition temperature of 650 C and 5 mTorr of oxygen [16]. As shown in Fig. 3, the as-deposited film was oxygen deficient, and low temperature annealing pushed the superconducting transition temperature over 80 K. The films made by this process were very smooth and shiny, and had a sharp interface with the substrate.

By simply increasing the oxygen pressure during deposition from 5 mTorr to 100-200 mTorr and using substrate temperature between 650-750 C, high quality superconducting thin films were obtained without further heat treatment after cooling down to room temperature in over 200 Torr of oxygen [17-21]. Higher oxygen pressure in the system enhances the formation of sub-oxides in the gas phase [22], and higher substrate temperature helps the film nucleation. A typical resistivity vs temperature is shown in Fig. 4 for a film deposited under those conditions. The film is highly metallic with a transition temperature and width of over 90 K and less than 1 K, respectively.

A 10 nm thick $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film on SrTiO_3 made by pulsed laser deposition had a transition temperature of 82 K [21], which is as good as those made by other techniques [23-24]. YBaCuO thin films made by laser deposition *in-situ* have one of the highest critical current densities ($\sim 5 \times 10^6 \text{ A/cm}^2$) at 77 K. Those films are also highly oriented with c-axis normal to the film surfaces, and crystalline with RBS channeling minimum yields of $\sim 3\%$ [25], which are as good as those obtained on bulk single crystals [26]. The films also exhibited the lowest flux noise measured to date [27], which further confirmed the high quality of the films. *In-situ* YBaCuO films on LaAlO_3 had an order of magnitude lower surface resistances compared to Cu at 77 K at a few GHz [25], showing promising future for microwave applications.

More recently, high quality YBaCuO superconducting thin films have been prepared *in-situ* by pulsed laser deposition at a deposition rate up to 14.5 nm/s [28], which is two orders of magnitude higher than the conventional rate of typically $\sim 0.1 \text{ nm/s}$. Those films had superconducting transition temperatures of 90 K. What is more remarkable is that the films at 77 K exhibited no marked degradation of current carrying capacity with

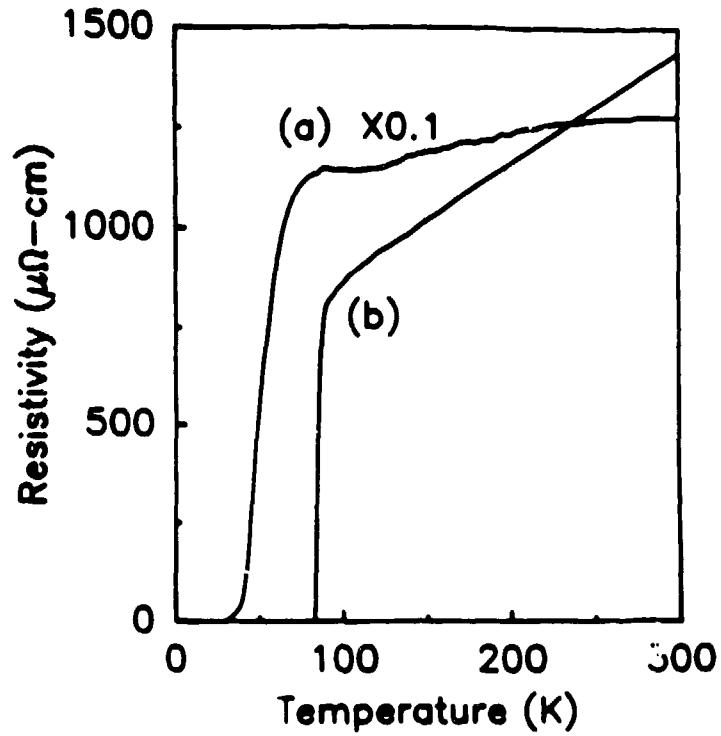


Fig. 3 Resistivity vs temperature for (a) an as-deposited YBaCuO film on (110) SrTiO₃ made at 650 C in 5 mTorr oxygen, and (b) the same film after annealing in oxygen at 450 C for 3.5 h.

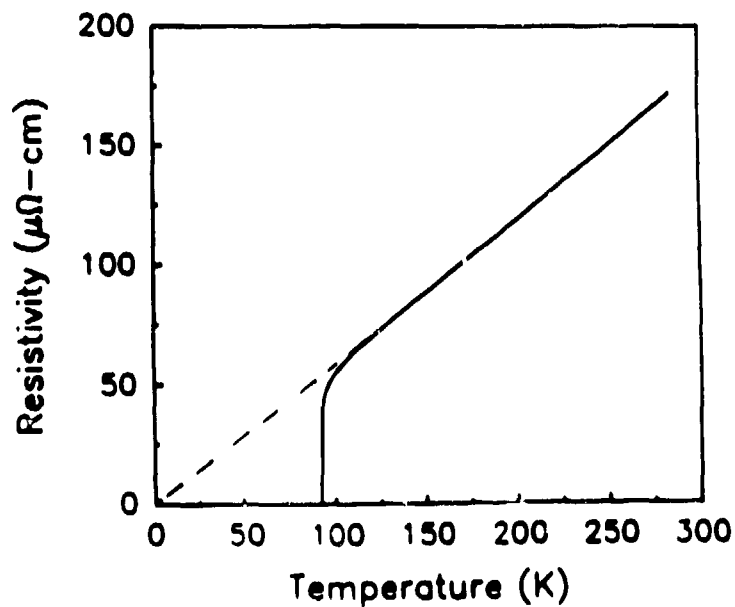


Fig. 4 Resistivity vs temperature for an as-deposited 100 nm YBaCuO film on SrTiO₃ made at 730 C in 200 mTorr oxygen.

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critical current densities of 4×10^6 A/cm² [29]. The high deposition rate achieved by the laser deposition enhances the efficiency of the technique.

HETEROSTRUCTURES AND SUPERLATTICES

In order to fabricate planar junction devices such as superconductor-normal metal-superconductor (SNS) or superconductor-insulator-superconductor (SIS), heterostructure systems are needed. Since the high T_c superconductors are crystalline materials, lattice matched multilayers are the only choice in order to have the superconductor maintain its transition temperature or order parameter right up to the interfaces. From the material growth point of view, the processing conditions should be very similar for all matched materials to assure the quality.

One of matched materials for YBaCuO is $Y_{1-y}Pr_yBa_2Cu_3O_{7-x}$ ($y=0$ to 1), which has an orthorhombic structure with nearly the same lattice constants over the entire alloying range [30]. YPrBaCuO thin films can be prepared at same processing conditions as YBaCuO films [31]. Most of the substrate materials can be also used as the matched systems.

Multi-layers were accomplished by laser deposition in-situ by employing a multi-target holder, which allows different targets to be placed in the laser beam. By alternating the targets, heterostructures were fabricated. Epitaxial growth of YBaCuO/PrBaCuO/YBaCuO trilayers [32] has been shown with the demonstration of ac and dc Josephson effects in the SNS junction devices fabricated from such trilayer structures [33]. 2.8 MeV He ion backscattering minimum yield of 6% on the YBaCuO top layer was observed, indicating high crystalline nature of the trilayers [32].

As increasing the number of the layers and reducing the individual layer thickness of the heterostructure, a superlattice including two or more different materials is formed. By fabrication of the superlattices, one will be able to form possible new metastable phases, and study the charge transfer effects due to alloying in the various sites, proximity effects at the interfaces, and so on. Moreover, fabrication of the superconductor superlattices in a controlled way may lead to layer by layer synthesis of high quality superconducting thin films.

Fabrication of multilayered Bi(Pb)-Sr-Ca-Cu-O thin films by laser deposition of successive layers was reported by Kanai et. al. [34]. More recently, excellent YBaCuO/DyBaCuO superlattices with the shortest periodicity of 2.4 nm have been made by DC magnetron sputtering [35]. Epitaxial YBaCuO/PrBaCuO superlattices with a similar periodicity were prepared by a pulsed laser deposition technique [36]. The composition oscillations in the superlattices (Fig. 5) and the presence of the satellite peaks in x-ray diffraction spectrum demonstrated the periodicity was achieved in the structures. High quality of the superlattices was confirmed by showing a 1.2 nm YBaCuO layer separated by 10 nm PrBaCuO layer, which is insulating at low temperatures, was still superconducting above 4 K [37]. The transport measurements on the superlattices revealed interesting physics [37].

DEPOSITION TEMPERATURES

At present, the deposition temperature needed for high quality superconducting thin films is still high (650-750 C) for all the thin film deposition techniques. Reducing the processing temperature is one of dreams for all the researchers. One of the ideas is to create more reactive oxygen by using an oxygen plasmas [38], and ozone [39]. Witanachchi et. al. [40] used a +300 V bias potential between the target and substrate during laser deposition to enhance the laser generated plasma plume. It was found that the substrate temperature could be reduced by 50 to 100 C to obtain same superconducting transition temperatures. Further experiment by Koren et. al. [41] using the atomic oxygen produced by photodissociation of N_2O also indicated the importance of the atomic oxygen. It should

be noted that the effect of atomic oxygen could be only seen at temperatures below 600 C [40-42], and the superconducting transition temperatures and critical current densities of thin films made under those conditions are not as good as those made at higher substrate temperature. Further research is still needed to reduce the processing temperature and at same time maintain the quality of the films.

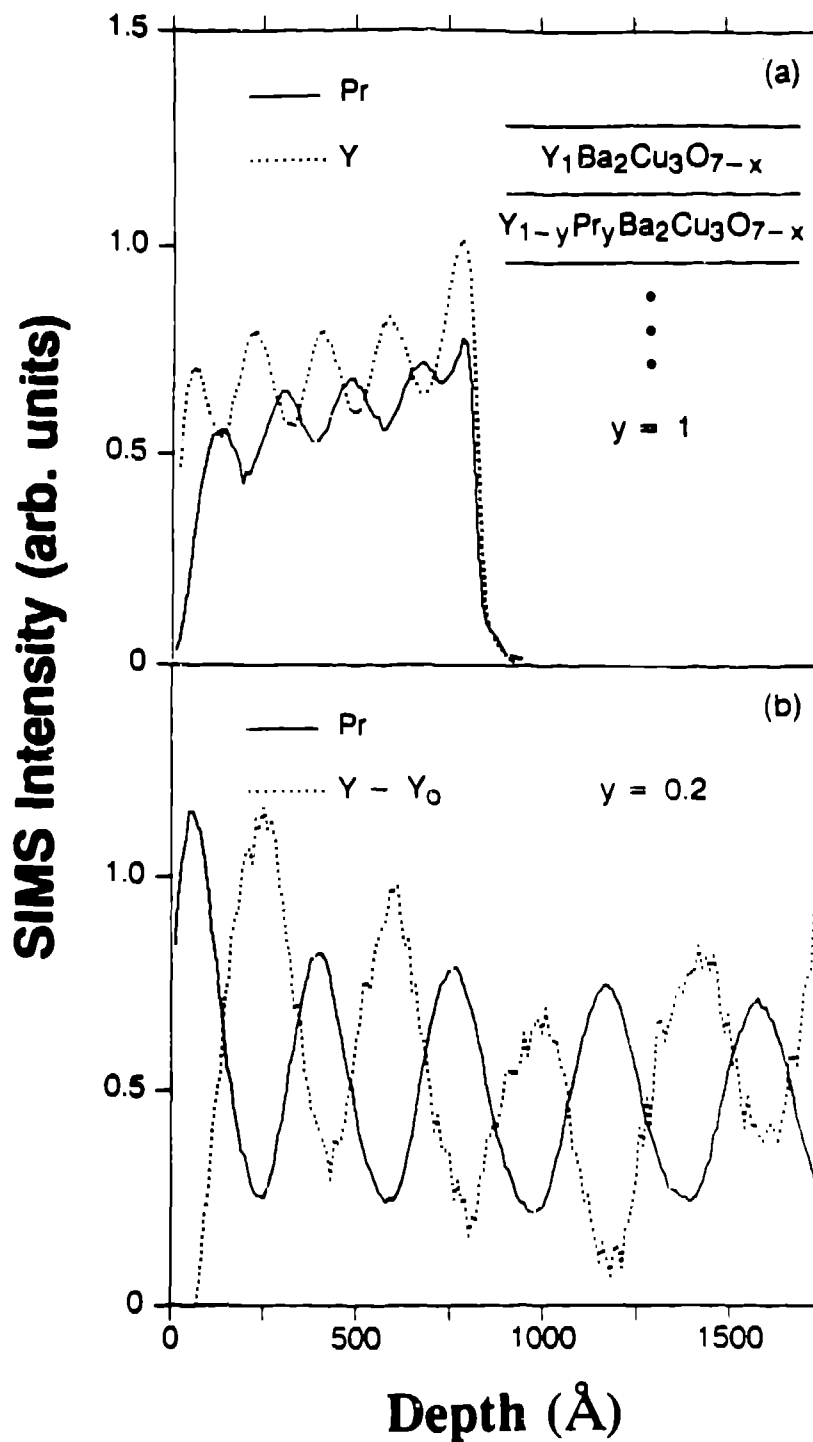


Fig. 5 SIMS data for two superlattices with different layer thickness: (a) 9 nm YBaCuO/9 nm PrBaCuO, and (b) 20 nm YBaCuO/20 nm $Y_{0.8}Pr_{0.2}BaCuO$. Y_0 is $\sim 75\%$ of the maximum Y signal. Y and Pr signals are exactly out of phase.

ADVANTAGES AND DISADVANTAGES

As mentioned above, the most important feature of pulsed laser deposition is the stoichiometric deposition. The use of a single target simplifies the deposition process [3]. Only single target sputtering such as inverted cylindrical magnetron sputtering [43] and off-axis sputtering [44] can compete with the pulsed laser deposition in these two areas. The versatility is an extra feature for the laser deposition. Simply shifting the targets, different materials or multi-layers are formed. The recent demonstration of high deposition rate using pulsed laser deposition is a plus. Another advantage of the technique is that a small amount of materials is enough as source materials and the target usage is much higher compared to sputtering.

To further demonstrate the advantages, a thin film was made from a small piece of Berlin Wall by pulsed laser deposition at room temperature in vacuum. Fig. 6 shows a RBS spectrum of the film on a carbon substrate, clearly indicating the existence of Ca, Si, Fe and O in the film. The composition of the film is close to the target average composition.

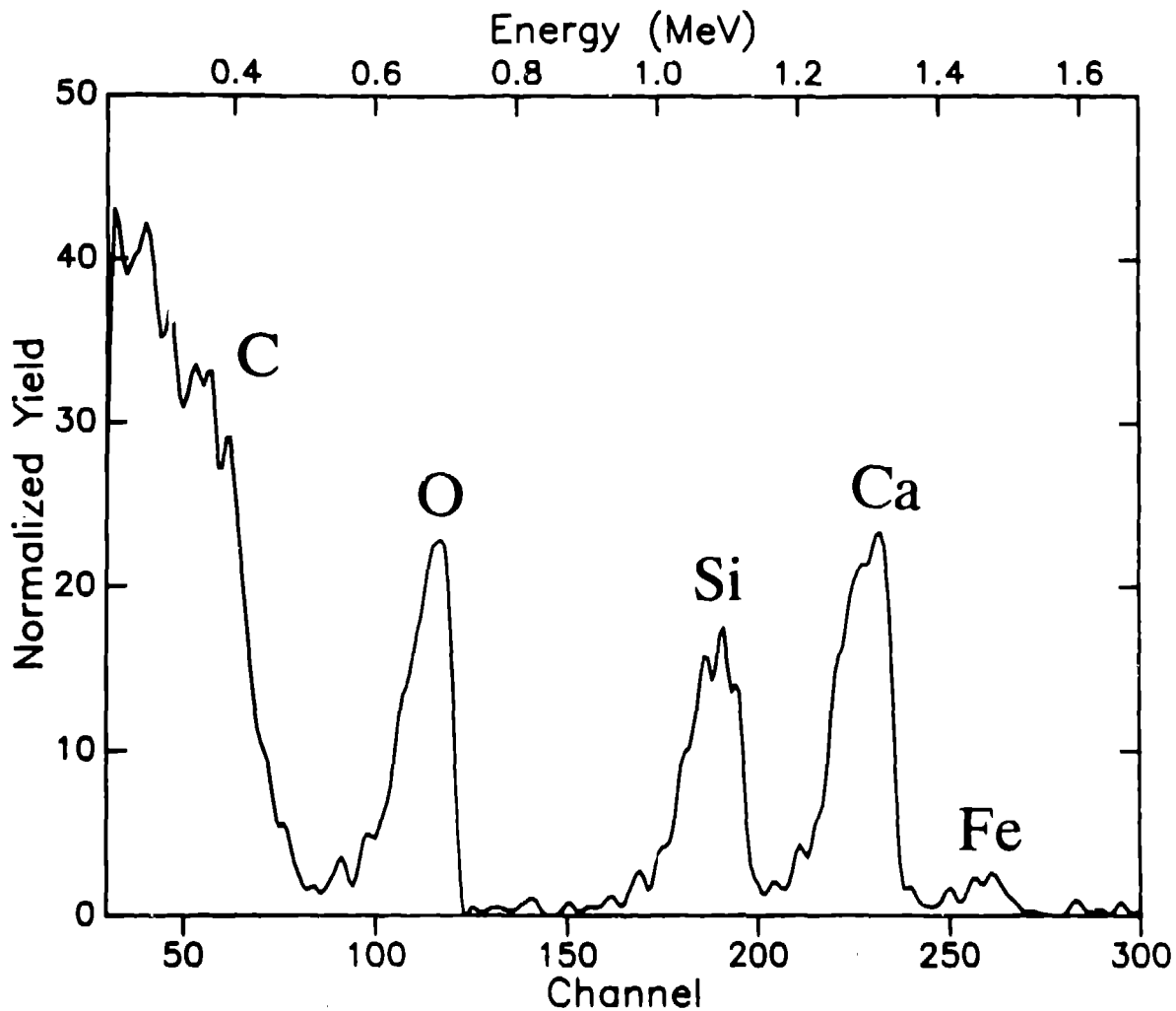


Fig. 6 A RBS (2 MeV He ions) spectrum of an as-deposited film on a carbon substrate made in vacuum from a small piece of Berlin Wall. The result indicates that laser deposition is a versatile thin film technique.

The main disadvantage of the technique is that the laser-solid interaction process is not fully understood at this time [3]. Much more work should be done in this area. Another problem is that there are always particles on the smooth surface of the films deposited by the laser technique. By using short wavelength lasers, the density and average size of the particles are relatively reduced [5]. For a single layer thin films, so far no measurements have shown that the existence of particles is degrading the superconducting properties of the films. Certainly, the particles should be removed in order to prevent possible shorting in the multilayers. The high cost of lasers is another concern.

One of the popular questions is whether the techniques can be scaled up. Since the deposition is highly forward-directed, one can image the laser deposition as a spray gun. By relative movement of the gun or the substrate holder, large area coating will be possible. It is believed that it is an engineering problem.

CONCLUSION

The successful use of pulsed laser deposition of high T_c superconducting thin films will bring more research in the technique. Detailed research is needed for further development of the technique. It is obvious that pulsed laser deposition will be one of the most important thin film techniques not only for research but also for manufacturing.

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