

Pulsed Laser Deposition: Thin Films in a Flash

FEATURE

by T. Venkatesan and Steven M. Green

Pulsed laser deposition offers a very fast route to prototyping any thin-film coating

Physicists first heard about a high-temperature superconductor (HTS) in 1986 at the Materials Research Society meeting in Boston. Just one decade later, we can use flashes of laser light to generate an atomic “spray” that can produce an HTS in a thin film that is only 1.2 nanometers thick. This technique—called pulsed laser deposition (PLD)—offers many potential applications, from integrated circuits and optoelectronics to micromechanics and medical implants.

The spring 1987 American Physical Society meeting in New York included a special session on HTS materials, and the lecture hall became too packed to accommodate the audience. The entire session—now known as the “Woodstock of physics”—was televised and projected outside the hall for the overflow crowd to watch. The main speakers were applauded—like rock stars—and the entire “concert” went on till the wee hours of the morning. Those meetings spurred considerable work on making HTS materials from thin films composed of multicomponent oxides, which consist of oxygen and a combination of cations, such as yttrium, barium, and copper. The endeavor looked very promising.

Film experiments

Soon after those exciting meetings, one of the authors of this article, T. Venkatesan, decided to use such a laser to evaporate a sample of stoichiometric $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO; where x represents the presence of some oxygen vacancies), deposit it as a film on a surface, and see if the film’s composition was the same as the original sample’s. In so-called equilibrium, or conventional, evapora-

tion processes, comparable temperatures develop throughout the entire target and the elements evaporate from the surface at different rates; barium would evaporate more quickly than copper and yttrium would be the hardest to evaporate—leading to a film that differs in composition from the target. Nevertheless, Venkatesan reasoned that an excimer laser should generate nonequilibrium evaporation, because it penetrates only a thin surface layer on the target for a few tens of nanoseconds. He thought that might cause the target’s composition to be preserved in the resulting film.

A few days later, Venkatesan and his graduate student Xindi Wu and postdoctoral researcher Dirk Dijkkamp used a pulsed laser in air to evaporate a target of YBCO, which was deposited on a carbon substrate for composition analysis. Surprisingly, the PLD process reproduced the target’s composition for all the cations in the film.

Although laser evaporation had been used previously for making a variety of films, the nonequilibrium nature of short-pulse laser heating had not been fully exploited or understood. The work by Venkatesan and his colleagues served as the first demonstration that the composition of a multicomponent film is preserved by using nonequilibrium surface heating generated by a pulsed ultraviolet laser. That discovery spawned an exciting new way of making multicomponent thin films. Today, PLD is one of the fastest growing thin-film processes in multicomponent films.

To appreciate PLD, one must understand the basic philosophy behind thin-film deposition. This process transports elements from one location to another by supplying energy to elements in a source, causing them to be transported to a surface to be coated. Ideally, such a process coats the surface with a pure film of the correct composition.

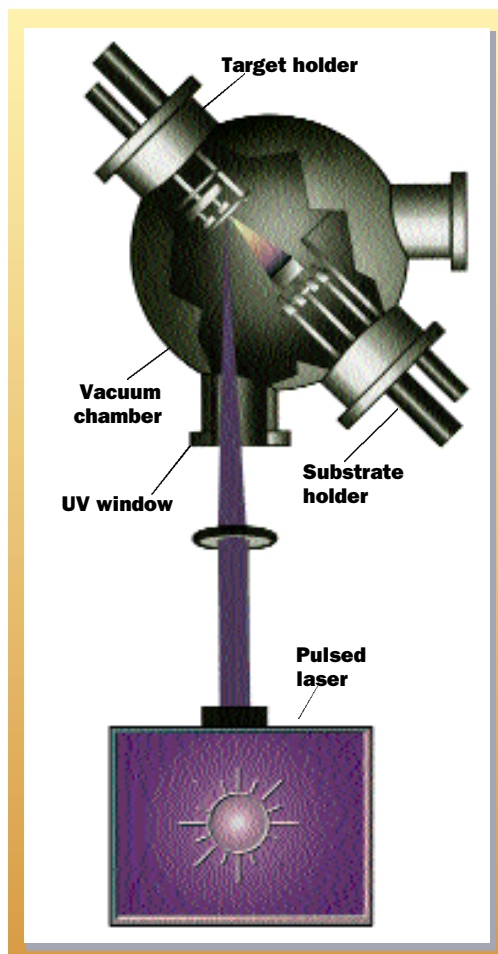


Figure 1. Pulsed laser deposition apparatus.

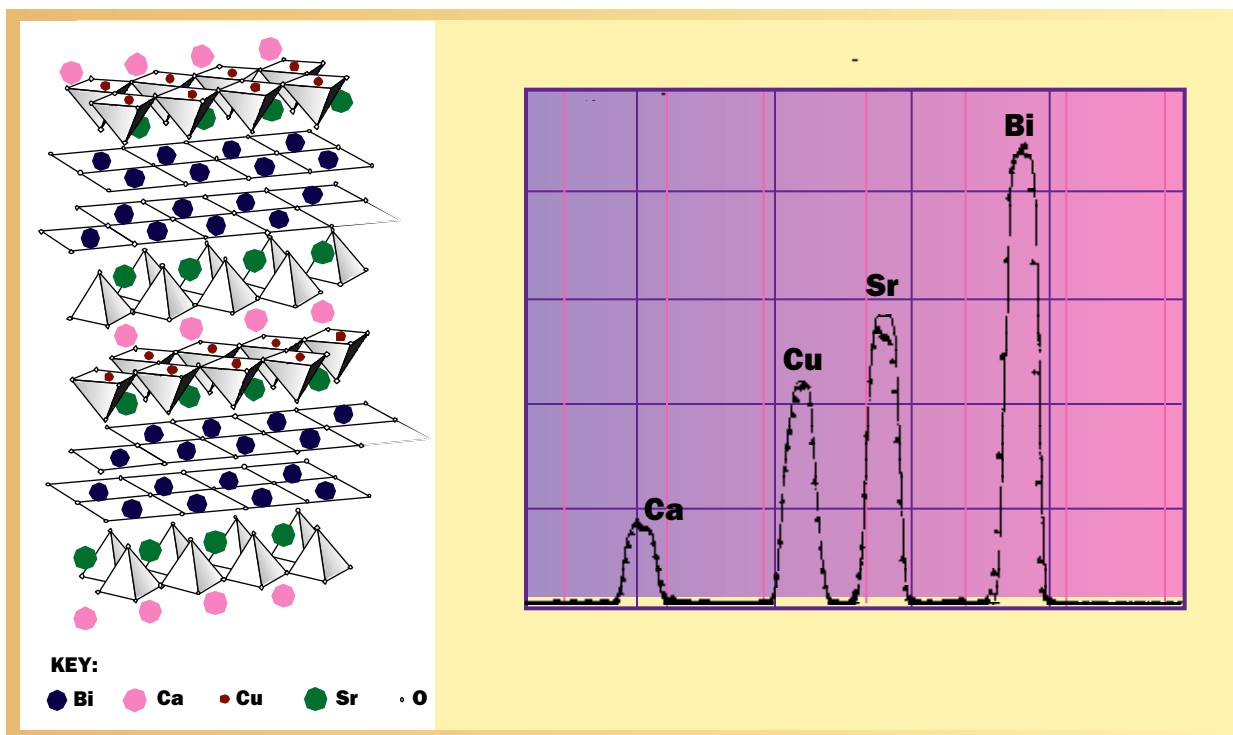


Figure 2.
Structure of $\text{Bi}_2\text{Sr}_2\text{Ca}_1\text{Cu}_2\text{O}_{10}$ target (left) and deposit's composition spectrum (right).

PLD's benefits

In PLD (Figure 1), a pulsed laser beam strikes the surface of the source material (also called the target), and the energy from the laser evaporates the target's surface. In most materials, the ultraviolet radiation is absorbed by only the outermost layers of the target—to a depth of about 1,000 Å. The extremely short laser pulses—each lasting less than 50 ns—cause the temperature of the surface to rise rapidly to thousands of degrees Celsius, but the bottom of the target remains virtually unheated—close to room temperature. Such nonequilibrium heating produces a flash of evaporants that deposit on the substrate, producing a film with composition identical to that of the target surface. Figure 2 shows the crystalline structure and compositional spectrum (as measured by the Rutherford backscattering technique)

of a rather complex multicomponent superconducting film in which the composition of the target is faithfully reproduced in the film. In Rutherford back scattering, the backscattering yield is proportional to the second power of the atomic number. The solid line in the figure is the expected yield for the target composition of ($\text{Bi}_2\text{Sr}_2\text{Ca}_1\text{Cu}_2\text{O}_{10}$), and the dots represent the

measured composition in the film. The ability to replicate the composition of the source in the film is perhaps the greatest benefit of PLD.

Several aspects of PLD are beneficial. It produces a highly forward-directed and confined plume of materials, which can be deposited with less contamination than, say, the unconfined plasma in a sputter process. In addition, PLD is incredibly precise. It can deposit a film of YBCO that is one unit cell (1.2 nm) thick; experiments show that a single unit cell of YBCO is a superconductor.

In complex multicomponent material deposition with conventional evaporation methods, the various cations come from different sources. To produce the right mixture in the deposited film, the rate of arrival of each species must be monitored and controlled. This

becomes especially difficult when large background gas pressures are used during the deposition. PLD does not require such monitoring, because the composition of the film replicates the composition of the target.

In most deposition techniques, the pressure of the background gas in the chamber puts severe limitations on the operating parameters. For electron-beam evaporation, the background gas pressure cannot exceed 10^{-4}

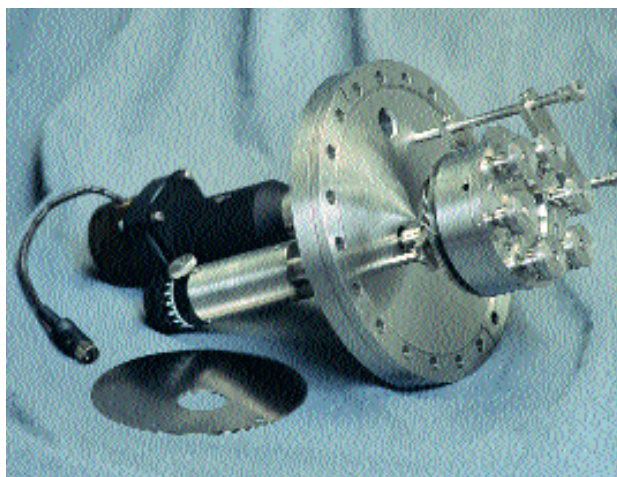
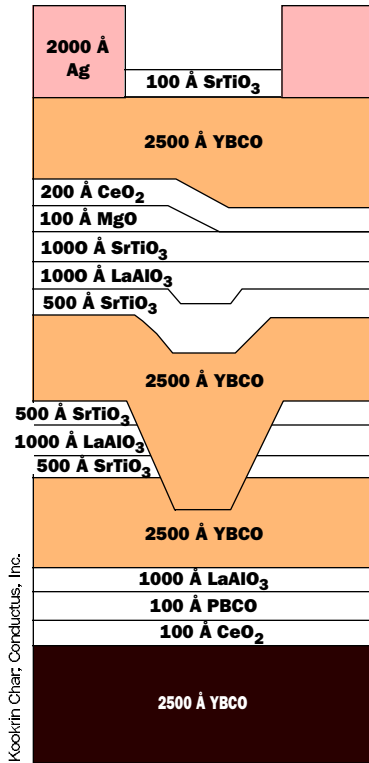


Figure 3. This target holder can position six different targets in the beam.



Kooklin Char, Conductus, Inc.

Figure 4. Fifteen-layer superconductor made with PLD at Conductus, Inc.

Torr, and for sputtering the pressure determines the rate of atom ejection from the target surface. In addition, different molecules require different background pressures for forming the correct phase. For instance, ZrO_2 films can be formed in vacuum, but $SrTiO_3$ requires an oxygen pressure of 50 mTorr. With PLD, the background gas pressure does not affect the passage or absorption of the laser, and the same system can be used to fabricate thin films composed of many materials by simply changing the background gas pressure. For example, PLD systems have been adapted to deposition systems in which the background gas pressure equals 1 Torr and to molecular beam epitaxy systems in which the pressure can be as low as 10^{-10} Torr.

Making multilayer materials can also be done rather easily with PLD, because different targets can be positioned under the laser beam. This is generally done with a computer-controlled target holder or carousel. Figure 3 shows a device that can position six different targets in the beam. Figure 4 shows a 15-layer superconducting device that was produced with PLD.

PLD is a good technique for depositing extremely pure films. In most processes, the film includes contaminants. For example, thermal and electron-beam evaporation use a container for the source material, and the fairly high temperatures needed to evaporate the source materials can also evaporate parts of the container, thereby contaminating the deposition process. In metal organic chemical vapor deposition (MOCVD), volatile organic molecules transport the desired cations from the source to the final destination, and fragments of the organic molecules can end up in the film.

Applications

Who uses PLD today? Investigators at the forefront of synthesizing novel thin films are the biggest users, because PLD offers the fastest route to prototyping any thin-film coating. In fact, many multicomponent materials produced with PLD promise valuable applications.

For example, barium strontium titanium oxide can be used in making very large scale integrated (VLSI) circuits, where the sizes of capacitors can be reduced dramatically. PLD-produced iron-oxide films may be used in magnetic recording. Optoelectronic applications can use thin films of lithium niobium oxide made with PLD. Many other PLD-produced thin films appear in devices for a variety of other technologies, including environmental sensors, micromechanical devices, light emitters, medical implants, and various coatings.

Despite the advantages of PLD, for certain applications one obstacle remains. In some materials, PLD produces fine particulates—chunks of target material ranging in size from microns down to submicron dimensions. Fortunately, a number of schemes will eliminate these particulates. A high-quality target minimizes the problem, and active and passive techniques have been developed to filter out the heavy particulates.

Some members of the thin-film community also believe that PLD faces a problem in scaling up. Nevertheless, this concern cannot be supported. With industrial excimer lasers available today, we estimate that deposition rates of more than 1 micron- cm^2/s are possible, and that is a commercially viable deposition rate. Some PLD systems can already deposit a thin film on an 8-inch wafer. Moreover, the demand for improved electronics based on metal-oxide or other multicomponent thin films should accelerate the development of commercial-scale PLD systems.

Recommended Reading

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