AIM

Association for Information and Image Management

1100 Wayne Avenue, Suite 1100
Silver Spring, Maryland 20910
301/587-8202

Centimeter

Inches

MANUFACTURED TO AIM STANDARDS
BY APPLIED IMAGE, INC.

1 of 1

Note: This is a preprint of a paper being submitted for publication. Contents of this paper should not be quoted nor referred to without permission of the author(s).

Growth of Multilayered Epitaxial Films by Pulsed Excimer Laser Ablation

1993 LEOS Annual Meeting
Short Wavelength and Gas Lasers Section

Douglas H. Lowndes

June 1993

"The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. DE-AC05-84OR21400. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes."

Solid State Division
Oak Ridge National Laboratory
P. O. Box 2008
Oak Ridge, Tennessee 37831-6056
managed by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-84ORR21400

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MASTER

1993 LEOS Annual Meeting
Short Wavelength and Gas Lasers Section

ABSTRACT

The characteristics of pulsed laser ablation for epitaxial film growth are reviewed. New developments in the growth of heteroepitaxial multilayers, stabilization of metastable phases, and growth of semiconductor alloys with continuously variable composition, are described.

Douglas H. Lowndes Phone: (615) 574 - 6306 FAX: (615) 574 - 4814

Solid State Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6056

The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. DE-AC05-84OR21400. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

GROWTH OF MULTILAYERED EPITAXIAL FILMS BY PULSED EXCIMER LASER ABLATION

Douglas H. Lowndes
Solid State Division, Oak Ridge National Laboratory,
Oak Ridge, TN 37831-6056

The characteristics of pulsed laser ablation for epitaxial film growth are reviewed. New developments in the growth of heteroepitaxial multilayers, stabilization of metastable phases, and growth of semiconductor alloys with continuously variable composition, are described.

The field of pulsed laser deposition (PLD) currently is undergoing rapid growth. Initial interest in PLD was triggered by the demonstration that high quality high-temperature superconductor (HTSc) films could be grown *in situ* by pulsed laser ablation (PLA) of a polycrystalline target.¹ This interest has been sustained and broadened by the concurrent development of high-power ultraviolet excimer lasers that are reliable and convenient to use, and by growing awareness of PLDs's advantages and even some unique capabilities for epitaxial film growth.

The major advantages that distinguish PLD from other film-growth methods are the following:^{2, 3}

- (1) stoichiometric (congruent) transfer of material from the ablation target to the substrate, provided that the laser energy density focused on the target is above the threshold for formation of a laser-generated plasma;
- (2) a natural capability for reactive deposition, since there is no need for electron beams, filaments, etc. within the deposition chamber; and,
- (3) the use of energetic species and a laser-generated plasma to enhance growth rates and reduce growth temperatures.

Part of the attraction of PLD film growth is that it is conceptually and experimentally relatively simple, though the physics of the PLA process is quite complex. A pulsed excimer laser beam (typical pulse duration ~ 10 -40 ns) is focused (energy density ~ 1 -5 J/cm²) through a UV-transmitting window onto a rotating target in a deposition chamber. Absorption of the focused laser energy produces a rapidly-expanding plasma that contains both ground-state and excited-state neutral atoms and ions, as well as electrons. These species undergo collisions in the high-density plasma region near the target, resulting in formation of a highly directional flow normal to the target surface with initial velocities $\geq 10^6$ cm/s. When ablation is carried out in a reactive atmosphere such as oxygen, very simple atomic oxides also are formed in the "plume" of ablated material. A shock front also may form as a result of collisions between the expanding plasma and the ambient gas molecules.⁴ This front propagates with gradually decreasing velocity toward the substrate heater, which typically is located 5-10 cm away. Collisions with ambient gas molecules moderate the initially high (~ 50 -500 eV) kinetic energies of ablated species, so that film-growth actually occurs with the substrate "bathed" in only a low-energy (~ 0.1 -1 eV) plasma.

PLA has two characteristics that most authors have regarded as limitations.^{2, 3} First, particles ranging in size from ~ 0.1 to ~ 10 μ m in diameter often are present on PLD films. Second, a focused laser beam produces a strongly forward-peaked angular distribution of ablated material, so that uniform-thickness films are obtained only over a small angular range in the forward direction. However, impressive progress has been made recently in overcoming both problems. The number and size of particles can be minimized through use of a short-wavelength (e.g. excimer) laser.² Laser beam scanning over the target and proper target preparation also are important, and a mechanical "velocity filter" may be useful in some cases.⁵ Highly uniform film

thickness can be obtained simply by introducing relative motion of the ablation plume and the substrate, e.g. by scanning the laser beam over the target or by rotating the substrate heater under a fixed, radially offset ablation plume.^{2, 3}

The possibility of scaling up the PLD process, to deposit uniform films over large areas, has been demonstrated, as have attractive deposition and epitaxial growth rates.⁶ Excimer lasers currently in the advanced testing stage will make possible epitaxial film growth at rates that are limited only by film-growth mechanisms themselves, over areas that are sufficiently large for some applications. Recent advances that include atomic layer-by-atomic layer construction of artificial phases,⁷ entirely in situ growth of heteroepitaxial HTSc/ceramic structures,⁸ and the development of at least an embryonic hybrid HTSc/semiconductor processing technology⁹ represent solid progress and give a hint of developments yet to come.

As a tool for exploratory thin-film materials research, PLD's chief characteristics are its enormous versatility and simplicity. Through PLD's use there has been a significant increase in the number of complex, multi-element materials that can be grown as epitaxial films. New PLD-based techniques for epitaxial growth also still are evolving. Two recent PLD developments with potential practical applications are:

- (1) the demonstration that metastable HTSc phases can be stabilized and grown as epitaxial films, while the corresponding bulk materials can be formed only under extreme conditions or not at all;^{10, 11} and,
- (2) development of a new method to grow compound semiconductor films with continuously variable composition, by ablating a single target of fixed composition through a variable-pressure ambient gas containing one of the film constituents. We have used this new technique to grow $\text{ZnSe}_{1-x}\text{S}_x$ films with $0 \leq x \leq 0.52$ (using a ZnSe target and H_2S gas), as well as epitaxial heterostructures that contain both abrupt and continuously graded compositional changes.¹²

This research was sponsored by the Division of Materials Sciences, U. S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc..

References

1. X. D. Wu et al., *Appl. Phys. Lett.* **52**, 754 (1988); A. Inam et al., *Appl. Phys. Lett.* **53**, 908 (1988).
2. D. H. Lowndes, *Eighth Inter. Summer School on Crystal Growth*, Palm Springs, CA, Aug. 9-15, 1992 (in press).
3. J. T. Cheung and H. Sankur, *CRC Crit. Rev. in Solid State and Mater. Sci.* **15**, 63 (1988).
4. D. B. Geohegan, *Appl. Phys. Lett.* **60**, 2732 (1992).
5. W. P. Barr, *J. Phys. E* **2**, 2 (1969).
6. X. D. Wu et al., *Appl. Phys. Lett.* **56**, 1481 (1990).
7. M. Kanai et al., *Appl. Phys. Lett.* **54**, 1802 (1989); *Appl. Phys. Lett.* **58**, 771 (1991).
8. L. P. Lee et al., *Appl. Phys. Lett.* **59**, 3051 (1991).
9. D. K. Fork et al., *Appl. Phys. Lett.* **57**, 2504 (1990); *Appl. Phys. Lett.* **58**, 2294 (1991).
10. D. P. Norton et al., *Phys. Rev. Lett.* **66**, 1537 (1991).
11. D. P. Norton et al., *Appl. Phys. Lett.* **62**, 1679 (1993).
12. J. W. McCamy and D. H. Lowndes, submitted to *Appl. Phys. Lett.*

DATE

FILMED

10 / 13 / 94

END