OPTICAL MULTICHANNEL IMAGING OF PULSED LASER DEPOSITION OF ZNO (POSTPRINT)

John G. Jones
AFRL/RXAN

AUGUST 2014
Interim Report

Distribution A. Approved for public release; distribution unlimited.
See additional restrictions described on inside pages

STINFO COPY

© 2014 IEEE
Using Government drawings, specifications, or other data included in this document for any purpose other than Government procurement does not in any way obligate the U.S. Government. The fact that the Government formulated or supplied the drawings, specifications, or other data does not license the holder or any other person or corporation; or convey any rights or permission to manufacture, use, or sell any patented invention that may relate to them.

This report was cleared for public release by the USAF 88th Air Base Wing (88 ABW) Public Affairs Office (PAO) and is available to the general public, including foreign nationals.

Copies may be obtained from the Defense Technical Information Center (DTIC) (http://www.dtic.mil).

AFRL-RX-WP-JA-2014-0186 HAS BEEN REVIEWED AND IS APPROVED FOR PUBLICATION IN ACCORDANCE WITH ASSIGNED DISTRIBUTION STATEMENT.

//Signature//
JOHN G. JONES
Nanoelectronic Materials Branch
Functional Materials Division

//Signature//
DIANA M. CARLIN, Chief
Nanoelectronic Materials Branch
Functional Materials Division

//Signature//
TIMOTHY J. BUNNING, Chief
Functional Materials Division
Materials and Manufacturing Directorate

This report is published in the interest of scientific and technical information exchange, and its publication does not constitute the Government’s approval or disapproval of its ideas or findings.
1. REPORT DATE (DD-MM-YYYY)  
August 2014

2. REPORT TYPE  
Interim

3. DATES COVERED (From – To)  
09 November 2009 – 20 July 2014

4. TITLE AND SUBTITLE  
OPTICAL MULTICHANNEL IMAGING OF PULSED LASER DEPOSITION OF ZNO (POSTPRINT)

5a. CONTRACT NUMBER  
In-House

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER  
62102F

5d. PROJECT NUMBER  
4347

5e. TASK NUMBER

5f. WORK UNIT NUMBER  
X041

6. AUTHOR(S)  
(see back)

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)  
(see back)

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)  
Air Force Research Laboratory  
Materials and Manufacturing Directorate  
Wright Patterson Air Force Base, OH 45433-7750  
Air Force Materiel Command  
United States Air Force

10. SPONSOR/MONITOR’S ACRONYM(S)  
AFRL/RXAN

11. SPONSOR/MONITOR’S REPORT NUMBER(S)  
AFRL-RX-WP-JA-2014-0186

12. DISTRIBUTION / AVAILABILITY STATEMENT  
Distribution A. Approved for public release; distribution unlimited. This report contains color.

13. SUPPLEMENTARY NOTES  
PA Case Number: 88ABW-2013-5264; Clearance Date: 12 December 2013. Journal article published in Plasma Science Volume 42, Issue 10, 2590-2591. © 2014 IEEE. The U.S. Government is joint author of the work and has the right to use, modify, reproduce, release, perform, display or disclose the work. The final publication is available at DOI: 10.1109/TPS.2014.2342159.

14. ABSTRACT  
Pulsed laser deposition is an efficient technique to obtain stoichiometric material transfer from target to substrate and has been used by researchers and in industry for depositing materials for use in applications ranging from hard coatings and superconductors to optical materials. The images detailed here will demonstrate the unique plume evolution that occurs and the high-speed ionic species, and slow-speed neutral and molecular species that travel from target material to substrate.

15. SUBJECT TERMS  
imaging, materials preparation, zinc oxide

16. SECURITY CLASSIFICATION OF:  
a. REPORT  
Unclassified

17. LIMITATION OF ABSTRACT  
SAR

18. NUMBER OF PAGES  
6

19a. NAME OF RESPONSIBLE PERSON (Monitor)  
John G. Jones

19b. TELEPHONE NUMBER (include area code)  
(937) 255-9106

Standard Form 298 (Rev. 2-89)  
Prescribed by ANSI Std. Z39-18
**6. AUTHOR(S)**

John G. Jones, Neil R. Murphy, and Rachel Jakubiak - Materials and Manufacturing Directorate, Air Force Research Laboratory, Functional Materials Division  
Lirong Sun - General Dynamics Information Technology

**7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)**

AFRL/RXAN  
Air Force Research Laboratory  
Materials and Manufacturing Directorate  
Wright-Patterson Air Force Base, OH 45433-7750

General Dynamics Information Technology  
5100 Springfield Road  
Dayton, Ohio 45431
Optical Multichannel Imaging of Pulsed Laser Deposition of ZnO

John G. Jones, Lirong Sun, Neil R. Murphy, and Rachel Jakubiak

Abstract—Pulsed laser deposition is an efficient technique to obtain stoichiometric material transfer from target to substrate and has been used by researchers and in industry for depositing materials for use in applications ranging from hard coatings and superconductors to optical materials. The images detailed here will demonstrate the unique plume evolution that occurs and the high-speed ionic species, and slow-speed neutral and molecular species that travel from target material to substrate.

Index Terms—Imaging, materials preparation, Zinc Oxide.

PULSED laser deposition (PLD) processes utilize a high vacuum chamber, laser, mirror arrangement with focusing optic, target material, and substrate material. The vacuum chamber laser entrance window needs to be optically transparent for the laser wavelength of interest, and the target material must be both highly absorptive to the laser wavelength of interest and mounted in such a manner as to point at the substrate. Typically substrates are rotated for uniformity of growth and target materials are rotated for minimization of coning and target damage. When the focused laser pulse strikes the target material, a plume is generated that is ejected orthogonally to the target material. By controlling the chamber pressure, laser energy, focus spot size, substrate temperature, and target material, a variety of material compositions and morphologies can be deposited, and in this case ZnO is demonstrated [1].

For this effort, the ZnO laser ablation target was 50-mm diameter, a Lambda Physik LPX 220 laser was used having a wavelength of 248 nm, pulse length of \( \sim 20 \) ns, energy setting of 350 mJ/pulse, and a resulting target fluence of \( \sim 90 \) J/cm\(^2\). The chamber pressure was 2 Pa of oxygen. With focal distances being typically on the order of 0.75 m, and adjustments to focus being in the range of few millimeters, it is easy to see that process optimization can be simplified with some sort of \textit{in situ} tool of characterization.

Plasma diagnostics using an optical multichannel analyzer (OMA) can be invaluable in trying to diagnose a new deposition setup or new material system. The OMA used for imaging is an Andor Istar intensified charge-coupled device camera with manual focusing optic and electronic synchronization to the laser pulsing. In addition, a high-speed mirror system, Cambridge Technology’s two-axis optical galvanometer mirror system with capacitance coupled feedback for high speed motion, was used to steer the laser beam. During each actual thin-film growth or deposition, the laser beam is steered to a different position on the laser target randomly to obtain uniformity of growth of the thin film, and also to minimize damage to the target. To obtain the images in Fig. 1, the field of view did not include the actual target material that would have blinded the camera to the weaker optical emission of interest, i.e., the 0–1-\( \mu \)s duration image [Fig. 1(a) and (c)].

PLD using a single laser pulse \( \sim 20 \) ns generates a plume that results for \( \sim 20 \) \( \mu \)s. Two subprocesses can describe the different stages: a plume is generated as a function of the laser interaction with the target surface, and the subsequent plume contains different components including ions, excited molecular and atomic species, and neutrals that both interact during flight and with the oxygen present at 2-Pa pressure. A plume is brighter at higher background gas pressures, but thermalizes quicker and therefore decays away faster, requiring a shorter substrate distance. At lower pressures, the plasma travels farther due to a lower thermalization rate, but potentially the desired films would be substoichiometric and not have the desired oxygen composition. Ideally, PLD provides for stoichiometric transfer of materials from target to substrate, but oxygen is always lost due to the vacuum and hence the need for an oxygen background pressure to provide oxygen for reaction.

With the target-laser interaction out of the field of view of the OMA, Fig. 1 shows an integrated plume image of a single laser pulse of ZnO from 0 to 20 \( \mu \)s, where the composite can be described by a summation of Fig. 1(a) and (b). Fig. 1(a) is expected to be dominantly the Zn\(^{+1}\), and Fig. 1(b) is the composite of all different species integrated between 1 and 20 \( \mu \)s, demonstrating that the first frame alone seems to contain a high speed component of ZnO PLD [2].

Comparing Fig. 1(c) and (d) clearly shows that the high speed component is effectively gone within \( \sim 1 \) \( \mu \)s, having a velocity of \( \sim 50 \) km/s. Fig. 1(d)–(j) shows plume shock front and progression in 1 \( \mu \)s steps with a speed of \( \sim 7 \) km/s. The species dominantly expected in these plume images are...
Fig. 1. Optical multichannel analyzer (time gated ICCD) images for 1-µs durations showing pulsed laser ablation plume evolution of ZnO at 2 Pa of oxygen. Intensity is shown in arbitrary units, where the most intense plume fluence is plotted in white and the least intense in blue. (a) and (b) Notice the sharp image feature showing the initial high velocity ionic species from the plume, which typically is otherwise not observable as a 2-D measurement (typically, an energy analysis probe would have to be moved through the plasma to measure the fluence at each point in a 1-D fashion). (a) PLD ZnO 0–1 µs. (b) PLD ZnO 1–20 µs. (c) PLD ZnO 0–1 µs. (d) PLD ZnO 1–2. (e) PLD ZnO 2–3 µs. (f) PLD ZnO 3–4 µs. (g) PLD ZnO 4–5 µs. (h) PLD ZnO 5–6 µs. (i) PLD ZnO 7–8 µs. (j) PLD ZnO 19–20 µs.

Zn* and ZnO* [2]. Certainly there will be O* generated as well. The high speed component shown in Fig. 1(a) and (c) is expected to have been dominantly the Zn+1 as it would have the highest velocity when compared with excited molecular and atomic species of ZnO [1], [2]. Nanotechnology applications for ZnO require the highest standards of deposition, and one of the quickest tools for understanding and improving the deposition process can be accomplished using an OMA [3].

REFERENCES