

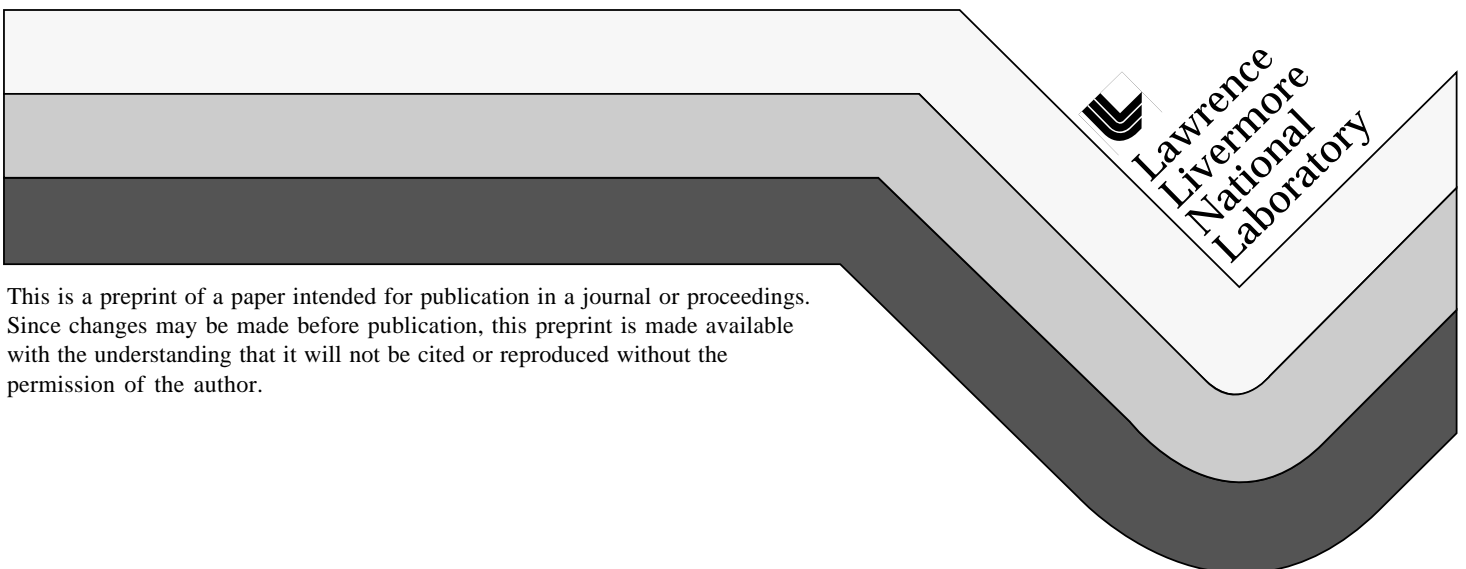
UCRL-JC-125736
PREPRINT

High Reflector Absorptance Measurements by the Surface Thermal Lensing Technique

Robert Chow
John R. Taylor
Z. L. Wu
Rene Krupka
Tianli Yang

This paper was prepared for submittal to the
Annual Symposium on Optical Materials for High Lasers
Boulder, CO
October 7-9, 1996

November 4, 1996



This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California 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 the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

High reflector absorptance measurements by the surface thermal lensing technique

Robert Chow, John R. Taylor
Lawrence Livermore National Laboratory
University of California, Livermore, CA 94550

Z. L. Wu, Rene Krupka, Tianli Yang
Department of Physics & Astronomy
Eastern Michigan University, Ypsilanti, MI 48197

ABSTRACT

Surface thermal lensing is an alternate configuration of a photothermal deflection system that was used to measure low levels of optical absorption. The thermal lensing configuration facilitated the alignment of the pump and probe laser beams by using a larger diameter probe beam. This technique was applied to high performance optical coatings, specifically high reflectors at 511 nm, zero degrees angle of incidence. The absorptance of these coatings was previously measured using a high power copper vapor laser system. A high power copper laser beam is focused onto a ~2 mm diameter spot. A thermal camera senses the temperature rise with respect to the rest of the coating. The temperature change, power density and beam diameter were used with an empirical formula that yields optical absorption. The surface thermal lensing technique was able to resolve absorption levels lower than that achieved with the copper laser method.

Keywords: weak absorption measurement, surface thermal lensing, photothermal radiometry, optical thin films

1. INTRODUCTION

High performance optical coatings for an atomic vapor laser isotope separation plant require simultaneous performance of a number of material properties, one of which is extremely low levels of absorptance. Low absorptance prevents thermal-induced distortion of the optic when exposed to high irradiances. Distortions affect the laser beam's wavefront and subsequently degrades the plant efficiencies. Low absorptance also reduces the incidence of laser-induced damage. The deposition process optimization cycle for absorptance involves a period of weeks as the coating supplier makes test runs, sends the coated witnesses to LLNL for absorptance testing, and uses the LLNL results to adjust the design or the process. The long time delays between test runs are not conducive to a manufacturing environment. A preferable method is to have an absorptance measurement instrument accessible to the coating supplier. This requires an instrument which is easy to operate, repeatable, reliable, and consists of commercially available laser and optical components.

The photothermal deflection (PTD) technique appears suited for this low absorptance application [1-3]. The technique has high sensitivity, uses a low to moderate power laser, and the coating may be tested at its design wavelength by selecting the appropriate pump laser. However, the technique involves the difficult steps of aligning two small diameter (~ 10 to 100 um) laser beams to close proximity in order to make the measurement. Coating-induced scatter makes the small beams appear larger so that critical positioning of the beams cannot be done visually. Also, the signal to noise ratio is low, requiring low-noise signal amplification between the detector and the lock-in amplifier. The surface thermal lensing (STL) configuration of the photothermal deformation technique is an alternate detection method [4,5] that has demonstrated more precision and easier alignment than the conventional set-up [6]. The reflected signal in an STL configuration has been modeled by diffraction theory, and demonstrated on high absorptance filter glass and low absorptance single layer refractive metal oxide layers [6]. The purpose of this work is to demonstrate that STL could measure absorptances on refractory metal oxide, multilayered coatings.

2. EXPERIMENT

Absorptance measurements by the PTD technique is illustrated in Figure 1. The absorption of the pump beam energy causes a corresponding local temperature rise of the sample, which leads to a local surface deformation due to thermal expansion. By probing the laser-induced surface deformation, whose magnitude along the perpendicular direction to the sample surface can be less than 0.1 nm, one can relate the detected signal to the optical and thermo-mechanical properties of the sample. Conventional PTD uses a second, tightly focused, laser beam (probe beam) on the deformed surface as the sensing method

(Fig. 1a). In the PTD set-up, the probe beam size is much smaller than that of the lateral dimension of the deformed area. Thus after reflection from the deformed surface, the probe beam experiences a change in direction which is proportional to the slope of the surface deformation. For the STL set-up, in contrast to PTD, a probe beam with a size similar to or larger than the lateral dimension of the thermally deformed area is reflected from the sample surface (Fig. 1b). The deformed area on the sample surface acts as a lens which diffracts the probe beam. The shape of the surface deformation is thus recorded in the diffraction pattern of the reflected probe beam, which can be analyzed by using either a CCD camera or a scanning photodetector.

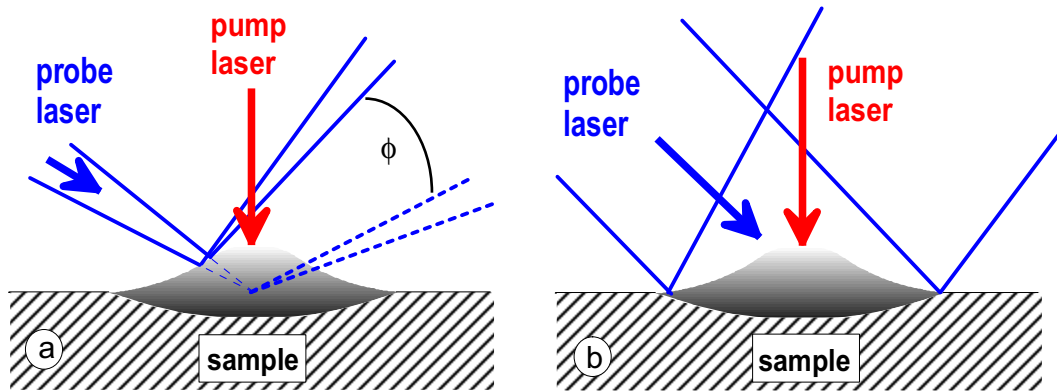


Figure 1 Comparison of a conventional photothermal deflection technique (PTD) and the surface thermal lensing (STL) method. The PTD requires two small diameter laser beams aligned close to each other. The probe beam diameter must be smaller than the lateral dimension of the thermally-induced deformation. The STL technique uses a probe beam diameter that is greater than the lateral dimension of the deformation.

STL offers two advantages over the conventional PTD configuration. STL has the same high sensitivity but avoids the critical alignment requirements of PTD. STL is easier to align because the probe beam is larger than the pump beam. The probe beam must overlap the pump beam, a diffraction-induced signal is obtained, and the signal may then be optimized. In the present work, both the pump and probe beam are in the visible region and observable to the naked eye. Furthermore, if a CCD camera is used for detecting the diffraction pattern, STL obtains the full field information of the surface deformation. This can be an important advantage over PTD, where the probe beam samples only a small spot of the deformed area. Less time is required to map the shape of the deformation.

The experimental setup for the absorbance measurements is illustrated in Figure 2. An Ar ion laser at 514.5 nm is used as the pump source. The beam is split so that the power can be measured, modulated at 12 Hz with a mechanical chopper wheel synchronized to a lock-in amplifier, and focused onto the sample at near normal angle of incidence with a beam diameter about 100 μm . The probe laser beam is from a 10 mW He-Ne laser, at the wavelength of 632.8 nm. It is focused onto the sample surface, coincident with the pump laser beam. For this particular set-up, the He-Ne laser beam diameter is focused to a diameter of about 500 μm to achieve optimum sensitivity of this set-up. The detector is located 60 cm from the sample. To detect the STL signal, a pinhole is placed in front of the photodetector. In this way the detector monitors intensity changes at the center of the probe beam, which is proportional to the optical absorbance of the sample. To convert the PTD signal into absorbance data, the traditional calibration method for PTD is used, where the calibration coefficient is achieved by measuring the PTD signal of a calibration thin film sample with known absorbance [2,3,7].

The coatings for the absorbance measurements are high reflectors designed to reflect wavelengths of 511 and 578 nm. All the coatings are made by a reactive e-beam process and deposited onto superpolished fused silica substrates. The substrates are 76.2 mm in diameter x 15 mm thick. The coatings are made by different manufacturers, and so have various material combinations and layer thicknesses.

The absorbance were measured by the STL technique described above and by a radiometric technique using a custom-built copper vapor laser (CVL) as the pump source [8]. The CVL is a high-average-power (200 watts), 4.4 kHz repetition rate, 50 ns pulse-width system [9]. The CVL beam is filtered to 511 nm light and focused approximately into a 2 mm diameter spot at 10 degrees angle of incidence onto the coating. An Inframetrics (model 525) infrared camera senses the temperature change with respect to the rest of the surface. The temperature change is converted into an absorbance value using an empirically

derived absorptance model [10]. This radiometric technique has been used successfully for the past decade to assist coating suppliers in the optimization of their respective coating processes and designs.

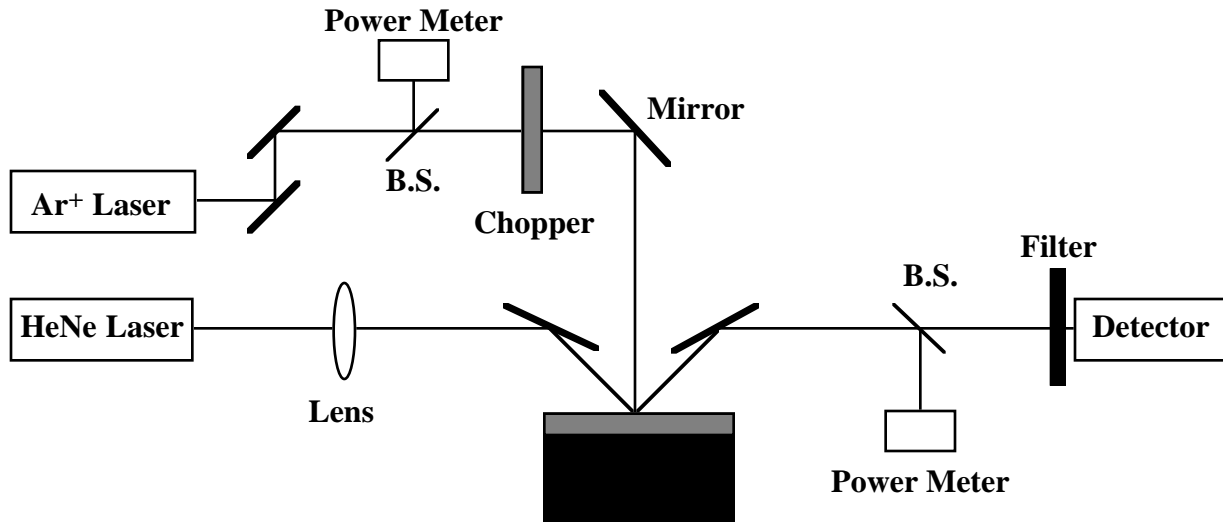


Figure 2. The surface thermal lensing set-up. The set-up is similar to that a conventional photothermal deformation set-up. The probe beam diameter at the surface is 500 μm , and the pump beam diameter is 100 μm .

3. RESULTS

3.1 Time Dependent Behavior

All the high reflector coatings exhibited a time-dependent absorptance behavior when measured with the STL technique. Four detailed time trends are shown in Figure 3. Note that each graphs plot absorptance as a function of time on different scales. The four time-dependent trends are:

- A. monotonic decrease with time; Most of the samples exhibited this particular behavior.
- B. step decrease; The initial absorptance is stable for about 60 sec but decreases approximately over a 100 sec time period to a lower, stable value.
- C. linear increase; This behavior is unacceptable for a high power laser coating and can lead to laser-induced damage.
- D. increase to a stable value.

Each graph is marked with an absorptance measurement for laser exposure times of < 10 seconds, 3 minutes, and > 4 minutes. The time-dependence behavior is well-established by exposing the coating for about 3 minutes.

The STL absorptances used in the Table 1 are the "long-term" absorptances taken at exposure times greater than 4 minutes. The data sampling rate is once per 3 sec until the coating absorptance appears stable. Then the sampling rate increases to once per 3 msec, and 250 data points are taken to determine an average and standard deviation of the stabilized absorptance value. The absorptance represents an average from 3 sites on the same sample.

The STL and the CVL-pumped technique show the same absorptance time dependencies for the samples, with only few exceptions. For SN 1352, an absorptance value of a few ppm is near the resolution limit of the CVL technique. The CVL-pumped technique is not able to resolve decreases of absorptance at this power density level. For SN 325, the sample was measured at two irradiances. The time trends match when the irradiances are both below 5 kW/cm². When the irradiance was higher, the time trends did not match. This may also be the reason why the time trends are not the same for SN 1289.

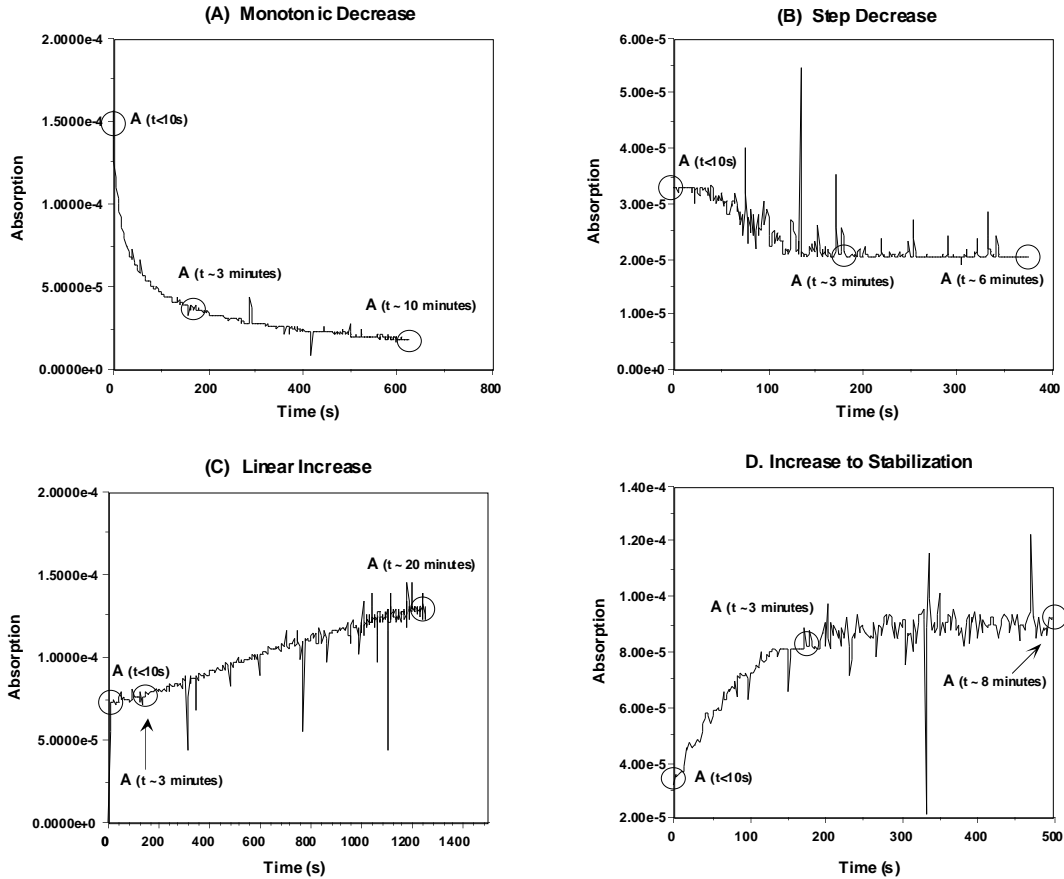


Figure 3 Absorbance time dependencies. Four cases of absorbance time dependencies are shown: (a) Monotonic decrease; (b) step decrease; (c) linear increase; and (d) increase to a stabilized value.

3.2 Nonlinear Behavior

In addition to time-dependent absorbance behavior, non-linear absorbance on SN 325 was observed as shown in Fig. 4. At two power irradiances, the stabilized absorbances are different. The absorbance at 8.7 kW/cm² is 500 ppm, and at 5 kW/cm², 133 ppm. Not only is non-linear absorbance behavior observed, but the time dependent behavior is different at the two irradiances. At the higher irradiance, the absorbance increase and begins to stabilize after 18 minutes. At the lower irradiance, the absorbance increases for the first 30 sec, and then decreases with time. The time trend at low irradiance is similar to that observed in Figure 3a. Over the long exposure time, the absorbance decreases monotonically with time. In the initial exposure, the absorbance increase is resolved on SN 325 but not with other coatings exhibiting a monotonic absorbance decrease.

The time- and power-dependent absorbance behaviors indicates that this information should be included along with the absorbance value. Specifically, along with the wavelength, polarization, angle-of-incidence, and pump-laser parameters, the absorbance value should include the irradiance of the measurement, exposure time to the laser beam, and the time trend. In fact, if the maximum service irradiance is known, the absorbance should be tested at this power density. This information will assist the end-users in determining the survivability of the optical coating in various applications.

Table 1 also shows a slight correlation between STL and CVL-pumped long-term absorbances. The data is sorted by CVL measurements from a low value of 2 ppm to high value 1500 ppm. The corresponding range for the STL values generally go from a low to a high value, but the range is much narrower, 6 to 500 ppm.

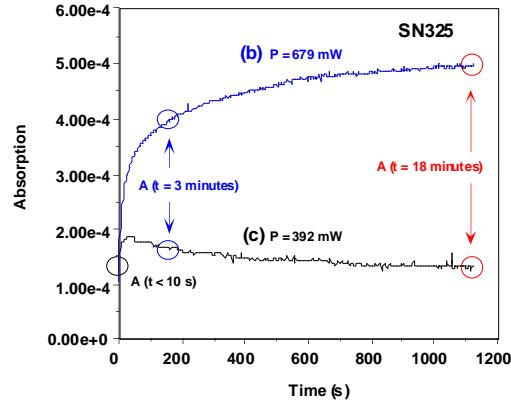
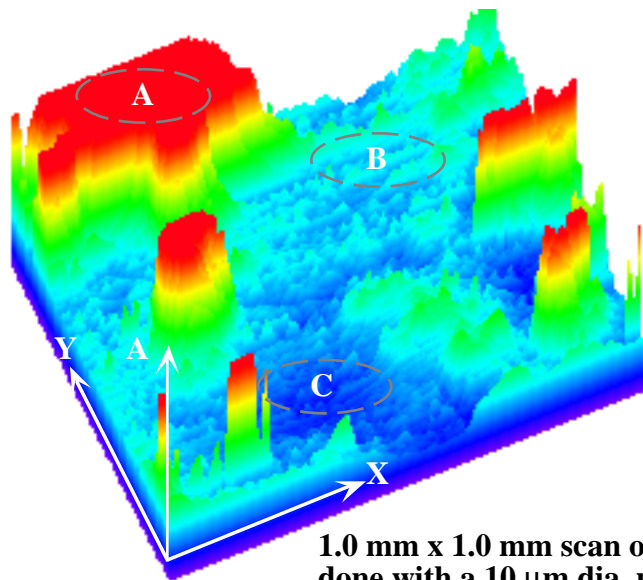


Figure 4 Non-linear absorptance. Sample SN 325 is measured at two different irradiances. The long term absorptance and the absorptance time-dependence is different.

4. DISCUSSION

Possible reasons that contribute to the low absorptance correlation between these two measurement techniques are presented. The absorptance is taken at different irradiances, as indicated in columns 3 and 5 of Table 1. The emissivity of the coatings is assumed constant for the CVL-pumped absorptance derivation. The pump lasers have different pulse modes (repetition rate and pulse widths) which could affect the absorptance values. The most significant contribution, we believe, is the spatial non-uniformity of absorptance in these coatings. Earlier work with Nd:YAG laser-pumped radiometry showed absorptance non-uniformity could be detected with beam spots < 0.45 mm in diameter [11,12]. In the present work, the beam diameters are about 2 mm and 0.10 mm for the CVL-pumped and the STL techniques, respectively. The STL technique is sampling an area about 400x time smaller than the CVL-pumped technique.

To demonstrate the degree of absorptance non-uniformity of a high reflector coating, a 2-dimensional scan was made on SN 1352 using the STL method. Figure 5 is a 3-dimensional scan of STL initial (< 10 sec of laser exposure) absorptances. The area is 1 mm x 1 mm and the pump beam diameter is 10 μ m to obtain an appropriate spatial resolution. An average of the whole area had a pixel value of 91 units; Area A had the highest averaged pixel value of 254 units; and area B had the lowest of 39 units. The intensity units translates proportionately into absorptance, meaning that the coating has a factor of 6 in absorptance variation in this area alone.



Sampling Area	Averaged Value	Dev.
Whole	90.79	57.69
Area A	254.00	0.00
Area B	38.74	3.67
Area C	68.06	7.15

1.0 mm x 1.0 mm scan of SN 1352 done with a 10 μ m dia. pump beam

Figure 5. Absorptance variation of a high reflector. The STL technique is used to measure the absorptance over a 1 mm^2 area of a high reflector coating. The pump beam diameter was decreased to $10 \mu\text{m}$ to increase the spatial resolution.

Absorptance results from a titania single layer are shown in Figure 6. A single layer is used that minimizes multilayer accentuating artifacts such as interfaces and process-induced defects. Figure 6 shows three scanned areas. There are two areas which were scanned with a high irradiance, 24 kW/cm^2 , and for dwell times about a minute per site. One area is square and labeled as $0.2 \times 0.2 \text{ mm}$. The other area is a rectangular and overlaps the square area. After the high irradiance scans were done, a low irradiance, 6.4 kW/cm^2 , scan was done over the area labeled $0.6 \text{ mm} \times 0.6 \text{ mm}$ at dwell times of less than a second per site. The absorptance is noticeably changed by the high irradiance/long dwell time scan, leaving a detectable footprint that is observable in the low irradiance/short dwell time procedure. Since a footprint is observed, the absorptance change lasts about 24 hours, the required time to make these measurements. However, the initial absorptance may return in time periods of a few months, as reported by reference [13]. The results also show that a PTD technique has sufficient stability to measure absorptances in a scan mode.

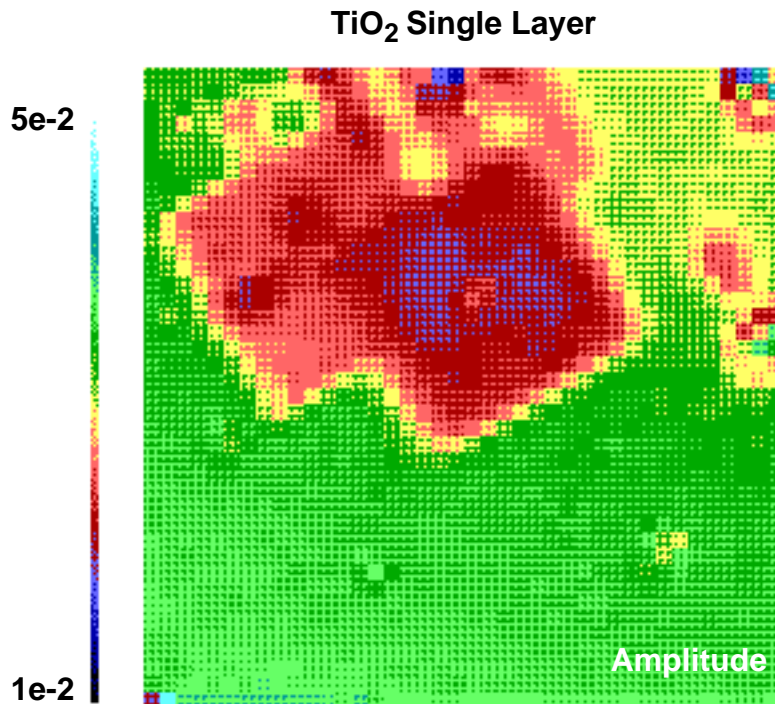


Figure 6. Three absorptance scans of the same sample. The two small areas ($0.2 \text{ mm} \times 0.2 \text{ mm}$) were scanned by high irradiance, which lowered the absorptances of this titania single layer through conditioning process. The absorptances were then detected by a subsequent low irradiance scan over a larger area (i.e. the whole picture, $0.6 \text{ mm} \times 0.6 \text{ mm}$).

5. CONCLUSIONS

The STL method has adequate resolution to measure low absorptances in high reflector, refractory metal oxide, multilayered coatings. A new protocol is suggested for reporting absorptance of high performance optical coatings. The reporting protocol should include

- i. the time dependence of the absorptance when the coating is under the maximum irradiation of the service application; an exposure time long enough to establish the time dependent behavior;
- ii. the irradiance, since coatings may have strong nonlinear or time dependent behavior at different irradiances;
- iii. the pump laser parameters (pulse modes, beam size, spectral requirements, etc.);
- iv. non-uniformity of coatings should be addressed if important for the applications.

6. ACKNOWLEDGMENTS

Auspices: This work was performed under the auspices of the U. S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48. Z. L. Wu at Eastern Michigan University acknowledges the support from the Faculty Research and Creative Project Fellowship and the State of Michigan Research Excellence Funds. The Equipment Grant (the LB-100 beam analyzer used in this work) from the Spiricon Inc. is gratefully appreciated.

7. REFERENCES

1. M. A. Olmstead, N. M. Amer, S. Kohn, D. Founire, and A. C. Boccara, "Photothermal displacement spectroscopy: an optical probe for solids and surfaces," *Appl. Phys. A* **32**, 141-154, 1983.
2. E. Welsh and D. Ristau, "Photothermal measurements on optical thin films", *Appl. Opt.* **34**, 7239-7253, 1995.
3. Z. L. Wu, M. Thompsen, P. K. Kuo, Y. S. Lu, C. J. Stolz, M. R. Kozlowski, "Overview of photothermal characterization of optical thin film coatings," in *Laser-induced damage in optical materials: 1995*, SPIE **2714**, 465-481, 1996.
4. P. K. Kuo and M. Munidasa, *Review of Progress in Quantitative NDE*, edited by D. O. Thompsen and D. E. Chimenti, Plenum New York, **8A**, 627-633, 1989; P. K. Kuo and M. Munidasa, "Single beam interferometry of a thermal bump," *Appl. Opt.* **29**, 5326, 1990.
5. H. Saito, M. Irikura, M. Haraguchi, and M. Fukui, "New type of photothermal spectroscopic technique," *Appl. Opt.* **31**, 2074, 1992.
6. Z. L. Wu, P. K. Kuo, Y. S. Lu, and S. T. Gu, "Laser induced thermal lensing for thin film characterization," in *Laser-induced damage in optical materials: 1995*, SPIE **2714**, 294-303, 1996.
7. Z. L. Wu, S. T. Gu, P. K. Kuo, Y. S. Lu, and Rene Krupka, "Non-destructive evaluation of thin film coatings using surface thermal lensing technique," to be published in *Thin Solid Films*, 1996
8. C. R. Stolz, J. R. Taylor, T. Sarginson, "Damage test capabilities using a high-repetition rate visible laser at LLNL," in *Laser-induced damage in optical materials: 1991*, SPIE **1624**, 109-115, 1992.
9. R. Hackel and B. Warner, "The copper-pumped dye laser system at LLNL," *SPIE OE/LASE'93*, Los Angeles, CA, Jan. 19, 1993.
10. V. G. Draggoo, R. G. Morton, R. H. Sawicki and H. D. Bissinger, "Optical coating absorption measurement for high power laser systems," in *High Power and Solid State Lasers*, W. Simmons, ed., SPIE **622**, 186-190, 1986.
11. A. F. Stewart, A. Rusek, and A. H. Guenther, "Thermal imaging studies of laser irradiated coated optical surfaces," in *Laser-induced damage in optical materials: 1988*, NIST Spec. Publ. **775**, 245-258, 1989.
12. S. E. Watkins, R. Heimlich, and R. R. Reis, Jr., "Mapping of absorption in optical coatings," in *Laser-induced damage in optical materials: 1991*, SPIE **1624**, 246-255.
13. C. J. Stolz, T. G. Sarginson, J. R. Taylor, "Non-linear absorption in high reflector multilayers," in *Laser-induced damage in optical materials: 1995*, SPIE **2114**, 433-442.

Technical Information Department • Lawrence Livermore National Laboratory
University of California • Livermore, California 94551

