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Laser modulated scattering as a nondestructive evaluation tool for optical surfaces and thin film coatings

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ABSTRACT

Laser modulated scattering (LMS) is introduced as a non-destructive evaluation tool for defect inspection and characterization of optical surfaces and thin film coatings. This technique is a scatter sensitive version of the well-known photothermal microscopy (PTM) technique. It allows simultaneous measurement of the DC and AC scattering signals of a probe laser beam from an optical surface. By comparison between the DC and AC scattering signals, one can differentiate absorptive defects from non-absorptive ones. This paper describes the principle of the LMS technique and the experimental setup, and illustrates examples on using LMS as a tool for nondestructive evaluation of high quality optics.

Keywords: laser modulated scattering, nondestructive evaluation, characterizations of defects, laser-induced damage, polished surfaces and thin film coatings

1. Introduction

Laser-induced damage in optical materials, whether these materials consist of optical coatings or bare substrates, is a localized phenomenon associated with the presence of micron and sub-micron scale defects [1-3]. These defects can be absorbers or non-absorbers. In the latter case the defects may cause field enhancement and/or reduction in heat conduction. Each, ultimately, can lead to materials damage.

A number of techniques have been introduced to investigate localized defects in optical materials. Among these techniques are scanning tools such as atomic force microscopy (AFM) [4, 5] and near field scanning optical microscope (NSOM) [6], and imaging tools including optical microscope and total internal reflection microscopy [7-8]. These tools, while having the capability of detecting many kinds of defects in optical materials, do not directly address absorption and thermo-mechanical response issues relevant to laser damage. In contrast, photothermal microscopy (PTM), based on optical beam deflection / diffraction [9] effect, has been developed as a tool for detecting optical absorption and thermal in homogeneity at the surface and inside the bulk of optical materials [10-13]. Using a low-power CW pump laser, PTM looks predominantly at linear absorbers [10-12]. Using a high power pulsed UV pump laser source it can probe multi photon absorption [13]. With an automated scanning system PTM has the ability to generate reproducible 2D photothermal images for both multilayer coatings and super polished fused silica surfaces [10 12]. For defects in bulk materials, PTM has been used to have the ability to generate reproducible 3D absorption maps for a KDP crystal [13].

The traditional PTM, while being useful for detecting micron-size and larger defects, has limited ability to detect sub-micron absorbers. For some optical materials the laser damage precursors are sub-100 nm in size [5,8,14,15]. The contribution of a small absorber to the overall photothermal signal can be overwhelmed by the background contribution from the host material. Furthermore, PTM as described above can not be used for studying damage growth because of the strong scattering from the damage site destroys the probe beam profile associated with the damage.

To complement the existing defect inspection/characterization techniques and overcome some of their limitations, a microscopic instrument has been developed that employs the principle of laser-modulated scattering (LMS). The technique allows simultaneous measurement of the scattering and the laser modulated scattering signal of a probe laser beam from an optical surface. Since no other parts of a super-polished optics but the defect sites generate scattering signal, the technique is a dark-field tool for defect detection on optical surfaces. By comparison between the scattering and LMS signals, one can differentiate absorptive defects from non-absorptive ones. Other
advantages of the LMS technique include its potential adaptability to lock-in imaging with focal array detectors and its high sensitivity to small defects even with large pump / probe beam sizes.

This paper briefly describes the LMS technique, summarizes the preliminary results that serve as a feasibility study, and discusses future applications of LMS to surface and subsurface defect inspection in optical materials.

2. Principle and model

The principle of laser modulated scattering from a defect is illustrated in Figure 1. For a typical microscopic tool (such as optical microscopy), it is the DC scattering from the defect that is measured, as shown in Figure 1 (a). If a pump laser is used to irradiate the defect, absorption at the defect and/or the host material will cause a localized temperature rise and hence a number of photothermal effects, including a change in the scattering field of the probe beam. By amplitude-modulating the pump beam, a modulated scattering field can be generated, as illustrated in Figure 1(b).

The modulated scatter, or LMS, signal can be detected using lock-in techniques. Mapping of an optic can be achieved by either scanning the sample or using a detector array. For the scanning case, the resolution is determined by the size of the pump and/or the probe laser beam. When imaging using a focal array detector, the pixel size of the image is the limiting factor for the spatial resolution.

Note that the spatial resolution should not be confused with the sensitivity of the technique for defect detection. The latter depends on the magnitude of the signal relative to the background, not the physical size of the defect. For microscopy based on LMS, the signal from a perfect surface is zero; therefore its sensitivity to local defects on or underneath a super-polished surface can be extremely high.

![Diagram of DC scattering and Laser modulated scattering](image)

Figure 1. Illustration of the principle of laser modulated scattering (LMS): (a). Scattering (DC) from a defect; (b). An amplitude-modulated pump laser beam is used to generate LMS.

The detection of light scattered by the laser-heated region is more informative than the DC scattering for laser damage studies, as long as local-absorption induced thermal / thermo-mechanical response remains the dominant damage mechanism. The variation of the refractive index due to laser heating typically is very small for optics with low optical absorption. Therefore the LMS signal can be described theoretically using a perturbation method, starting with a solution of the localized temperature rise caused by the laser heating.

Consider a Gaussian laser beam with a radius of a normally incident on the surface of the sample. Let us assume the contribution of the defect absorption is equivalent to a surface absorption of the local area with absorption coefficient $\alpha$. The localized surface temperature $T(t, r)$ is then given by formula [16]

$$T(t, r) = \frac{\alpha p^2}{k\sqrt{\pi DT}} \int_0^t \frac{t \left( \frac{t}{T} - \tau \right)}{\sqrt{\tau (p^2 + \tau)}} \exp\left(-\frac{r^2 p^2}{(p^2 + \tau)}\right) d\tau$$

(1)
Where \( t \) is time, \( T \) the pulse length of the mechanical chopped pump light, \( r \) the distance from beam center, \( a \) the pump beam radius, \( \alpha \) the thermal conductivity, and \( D \) the thermal diffusivity of the sample.

Equation (1) is applicable when the absorption depth is smaller than the thermal diffusion length. It is also applicable if the absorption takes place in few small subsurface defects within the laser beam.

The laser-induced temperature rise can be detected using LMS with different detection schemes. For the pump-probe detection scheme as shown in Figure 1 the scattered signal of the probe cw beam contains frequency components different from the modulated pump beam. In this paper the pump laser beam is mechanically chopped, i.e. it is a train of rectangular pulses with intensity \( I \), pulse length \( T \) and with interval \( T \) between the pulses (50% duty cycle). In the case that the probe beam size is much larger than the pump beam size, Equation (1) can be simplified [16] and the Fourier harmonic of the temperature field is given by

\[
T_0 = -\frac{i\alpha I a^2}{k \omega T r}\frac{\sin^2 \frac{\omega T}{2}}{1 + p(1 + i)} = \frac{\alpha I a^2}{2D} \tag{2}
\]

When the lock-in technique is used to detect the first harmonic signal of LMS, the signal represents the first harmonic temperature rise as follows

\[
T_1 = -\frac{i\alpha I a^2}{k \omega r(1 + p(1 + i))} = \frac{\pi a^2}{2DT} \tag{3}
\]

Figure 2. The LMS signal (amplitude) as a function of the product of DC scattering of the probe beam and local absorption of the pump beam, calculated assuming a constant incident pump laser power and a fused silica sample.

From Equation (3) it is apparent that the amplitude and phase of the LMS signal contain information on optical absorption, thermal diffusivity, as well as the location of the defect relative to the heating beam. Figure 2 shows the calculated dependence of the LMS signal as a function of the product of scattering and local absorption. The linear relationship gives us confidence of the technique to detect localized absorption, provided that the LMS signal can be normalized to the scattering signal of the probe laser beam. It also shows that for a defect that is both absorptive and scattering (e.g. a contaminated micro-crack or an absorptive metal inclusion) the LMS signal is particularly enhanced.
Figure 3 shows the calculated result of the LMS signal as a function of the thermal conductivity of the host material, assuming a spherical absorptive defect inside a fused silica sample but adjacent to its surface. Both the amplitude and phase of the LMS signal are strongly influenced by the thermal conductivity of the sample, demonstrating the sensitivity of the technique to thermal properties. It should be pointed out, however, that the current model is not sophisticated enough to quantitatively define the effect of localized thermal properties. For that purpose a rigorous model is needed based on the specific geometry of the defect and its relation to the host material. Work towards that direction is currently in progress.

![Graph showing LMS amplitude and phase as a function of thermal conductivity and modulation frequency.](image)

Figure 3. Calculated result of the LMS amplitude (a) and phase (b) as a function of the thermal conductivity of the host material, spherical absorptive defect inside a fused silica sample but adjacent to its surface.

3. Results and discussion

3.1 LMS signal as a function of the pump laser power

Optical scattering from a small defect / particle in general is a complicated phenomenon. The scattering signal measured by a detector is dependent not only on the size, shape, and properties of the scatter, but also the properties of the incident laser (wavelength, polarization, and incidence angle) and the position and size of the detector. Therefore, optimistically one says that scattering is a useful tool for defect characterization, and pessimistically one says that scattering is too complicated to be meaningful. As a result of its complexity and potential, scattering has been intensively and extensively studied and has been widely used for defect characterization and particle sizing [17].
The understanding of LMS is further complicated by the transient nature of photothermal response of an unknown defect and the resulting modification to the scattering field. While a rigorous model of LMS is under development, experimentally it is found that the amplitude of the LMS signal is proportional to the pump laser energy absorbed by the sample when the pump laser power is at appropriate levels. When the pump power goes to high levels, nonlinear response may dominate the LMS signal. The specific level of the threshold of nonlinear behavior differs from sample to sample. Figure 4 shows the relationship between the LMS amplitude signal and pump laser power from a defect on the surface of polished laser glass. The threshold of the nonlinear behavior in this case is at the level of about 1 kW/cm².

Figure 4. LMS signal as a function of the pump laser power. The sample used is a defect on the surface of laser glass. Experimental parameters: pump laser wavelength 488 nm, probe beam wavelength 633 nm, chopping frequency 70 Hz, pump beam and probe beam sizes ~100 μm.

The proportionality of LMS signal to absorbed energy at low power levels is observed for a variety of samples, including contamination particles on optical coatings and defect sites on the surface of polished laser glass. It shows that the LMS signal is proportional to the level of energy absorbed, as predicted by the model in section 2. Therefore, scanning an optical surface by using a constant laser power maps absorption of the surface if the LMS signal is normalized to the DC scatter signal.

3.2 LMS as a tool for characterization of coating defects

LMS has been applied to a variety of low absorptive optical components. Figure 5 shows a typical result for a low absorptive multilayer optical coating obtained by using (a) LMS and (b) DC scattering mapping. The images are taken from the same area, with an imaging size of 1 mm² and a spatial resolution of about 10 μm. The lines drawn in the images are for eye guidance when comparing the two images. It is found that LMS and DC scattering maps have only a weak correlation. For example, the absorptive defects C, E, I, J found using LMS do not show up at the DC scattering map, and the DC scattering defect K is not observed using LMS. Further, in the DC scattering map defect A has the highest amplitude, but the LMS result shows that defects B, D, G are as absorptive. We might therefore expect that B, D, G are also highly susceptible to laser damage even though they have weak scattering signals.

The difference in the sensitivity of the DC and AC signals is further shown in Figure 6 showing the profile of defect D shown in Figure 4. Compared with the signal from the background material, the DC scattering of the defect is only 2.8% higher but the LMS signal is about 10 times higher, showing that it is a strongly absorptive defect and a probable laser damage precursor.
Figure 5. Defect mapping using (a) LMS and (b) DC scattering of the same area of an optical coating (pump wavelength 1.06 µm; probe beam 0.6328 µm; pump beam size ~5 µm; probe beam ~25 µm; pixel size ~10 µm).

Figure 6. Profile of defect D shown in Figure 5. Compared with the background material, the DC scattering of the defect is only 2.8% higher but the LMS signal is about 10 times higher.

3.3 LMS as a tool for laser damage site characterization

The growth dynamics and mechanisms of a laser damage site under subsequent laser shots are influenced by the optical and thermo mechanical properties of the damaged site and the host material. Quantitative nondestructive evaluation (NDE) tools for damage sites are largely unavailable, other than topographic techniques such as SEM. Figure 7 shows results from the use of LMS to study laser damage sites on multilayer optical coatings. From the LMS amplitude image it is found that the damage site has a photothermal value about 16 times higher than the non-damage area. The enhanced photothermal signal at the damage site can be due to either a change of coating structures at the damage site or a physical modification of the materials or both. It is an indication of enhanced absorption and will lead to increase in absorbed energy from the subsequent laser shots, and very possibly, growth of the damage site.

The LMS phase image of the same damage site shows that it has a low thermal conductivity / high thermal resistance near the center, as demonstrated by an almost 180 degree phase change. The size of the thermal
inhomogeneity is much smaller than the absorption site shown in the amplitude map. This thermal inhomogeneity can be due to laser-induced delamination, micro-cracks, and/or removal of the coating materials at the center of the damage site.

The above results show that LMS can be a useful tool to non-destructively evaluate (NDE) damage sites and potentially correlate their properties with laser damage growth dynamics. Implementation of the technique into a damage testing system for in-situ studies may therefore be useful in the study of damage growth mechanisms.

![Figure 7. LMS image (left: amplitude; right: phase) for a laser damage site of an optical coating sample. The defect labeled as C is laser-induced debris.](image)

3.4 LMS as a tool for contamination studies

High power laser optics can be contaminated by improper handling, contaminated use areas, or by debris resulting from laser damage. A diagnostic tool that detects and characterizes contaminants is therefore of interest to laser damage studies. Figure 8 shows an example for such applications, where the laser-induced debris (caused by the damage shown in Figure 7) is scanned with micron spatial resolution. Both the amplitude and phase images indicate that the debris consists of two separated parts, profiles of which are shown in Figure 9.

From the image as well as the profiles, a few comments can be made about contaminants A and B. First, both are absorptive, with absorption more than 7 times higher than the background host materials.

Second, both of them are not well contacted to the background material, as can be seen from the phase image. The phase at the center of contaminant A is about 180 degrees different from that of the background. The phase jump happens at the edge (Profile A2-A2), spatially corresponding to the dip in the amplitude signal (Profile A1-A1). The combination of the amplitude and phase signals suggests a thermal-wave interference phenomenon caused by the poor thermal contact between contaminant A and the host material.

Third, the LMS image for contaminant B is not as symmetric as that for A. This asymmetry is observed more clearly in the profiles shown in Figure 9. The phase difference exists only for the right half of contaminant B (Profile B2-B2), corresponding also to the dip in the amplitude signal (Profile B1-B1). The results indicate that contaminant B is better contacted to the background material, with thermal resistance present only for a small portion of the interface. The contact between the contaminant and the surface is relevant to laser cleaning, conditioning and damage processes.

The above interpretations of the data need to be further verified by laser damage testing as well as a quantitative modeling of the LMS signals. Nevertheless, the results have demonstrated the potential of LMS as a quantitative NDE tool for detection and characterization of contaminants.
Figure 8. High resolution LMS image of the laser induced debris as labeled as C in Figure 7. The amplitude and phase images indicate that the debris consists of two parts, i.e., A and B; profiles of both parts are shown in Fig. 9.

Figure 9. Profiles of Section A and B of the debris as shown in Figure 8. While for both sections the signal peaks at the center, part A and B differs in that A is more symmetric than B. Further discussions can be found in the text, which suggest that part A has a poor contact with the sample surface while part B is well attached to the surface.

4. Summary

LMS is demonstrated to be a sensitive and non-destructive evaluation tool for defect detection and characterization of optics for high power laser applications. Results from optical coating studies show that the technique is also a promising tool for damage growth and contamination studies. Compared with existing techniques for defect characterization, LMS has a few distinguishing features that warrant wide application of the technique. First and
foremost, for super polished optical surfaces LMS is a dark-field tool and hence has higher sensitivity for small
defect detection than conventional PTM techniques. Second, by detecting DC scatter and LMS signals
simultaneously the technique is sensitive to both absorptive and non-absorptive defects and can separate them from
each other. Third, by analyzing LMS phase signals thermal inhomogeneities can be detected. Research towards
this direction need a more sophisticated modeling effort, which in currently in progress.

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