

# Photothermal absorption microscopy of defects in $\text{ZrO}_2$ and $\text{MgF}_2$ single-layer films

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**Abstract.** Photothermal displacement microscopy has been used for the characterization of  $\text{ZrO}_2$  and  $\text{MgF}_2$  single-layer thin films to detect absorption inhomogeneities at  $\lambda = 514$  nm. Images are presented for films of  $\lambda/2$ ,  $\lambda$ , and  $2\lambda$  optical thickness deposited on BK7 glass and SQ1 quartz substrates. Applying modulation frequencies ranging from 1 to 100 kHz a lateral resolution of several micrometers was obtained. We found that the size and density of the absorption inhomogeneities as well as absorptance depend strongly on the deposition process and weakly on the substrate material. No dependence on thin film thickness was found. The apparent defect density varies with the modulation frequency demonstrating the capability of the photothermal method to localize absorption in various depths of the thin film. Defect densities derived from the photothermal measurements are compared with results from total integrated light scattering (TIS) experiments performed on various samples at  $\lambda = 632$  nm. TIS intensities for  $\text{MgF}_2$  films on glass substrates were about one order of magnitude smaller than those from films on quartz. The latter revealed strong long-range (1-mm) variations of the light scattering intensity. This finding is in accordance with the absorption measurements.

*Subject terms:* photothermal microscopy; thin film systems; defect densities.

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## 1 Introduction

For the fabrication of optical thin film systems a detailed knowledge about residual absorption is essential. Such information helps to improve the optical quality of components. Also, the damage resistance of thin films for high-power laser applications may be increased substantially. Kozlowski et al.<sup>1,2</sup> and Staggs et al. recently presented clear evidence for a correlation between the laser-induced damage threshold and the presence of nodular defects in high reflectivity coatings at 1.06  $\mu\text{m}$ . Atomic force microscope (AFM) studies<sup>3</sup> revealed that the most severe coating quality degradation results from micrometer-sized defects distributed randomly over the surface with a density of approximately 10 per squared millimeter. It has been shown previously that the damage resistance of coatings can be improved by laser conditioning,<sup>4-6</sup> resulting in structural changes of the nodule defect morphology. Residual absorption is also a particular relevance for applications involving UV lasers or ultrashort light pulses. In these cases, the damage mechanism is often determined by direct bond breaking following the electronic excitation, whereas thermal processes play a minor role.<sup>7</sup> On the other hand, subthreshold irradiation with a focused laser beam can

lead to incubation in optical thin films. Recently, such a phenomenon has been demonstrated for oxide films where 14-ns excimer laser pulses at 248 nm generate regions with a high density of absorption centers.<sup>8</sup>

For a detailed investigation and control of all these phenomena and processes *in situ* absorption monitoring techniques are sought that provide both ultrahigh sensitivity as well as a spatial resolution in the micrometer range.

For absorption monitoring in thin films, a variety of conventional techniques have been applied. Absorption has been determined indirectly from measurements of optical transmittance and light scattering. Direct measurements of absorption were performed by calorimetry.<sup>9,10</sup> The main drawback of these conceptually and practically straightforward approaches is the limited sensitivity obtained. Especially for advanced optical materials that have been optimized for maximum transmission or reflection, a reliable characterization of absorption is not possible with these conventional methods.

Advanced absorption monitoring should aim at increased sensitivity as well as providing spatial resolution. We show in this paper that the average absorption measured by techniques with low spatial resolution is often an integrated effect of localized absorption centers rather than caused by a homogeneous absorption over the entire film. Our results are in accordance with the previously mentioned AFM studies.<sup>1-3</sup> Therefore, it is desirable to develop measurement systems capable of absorption measurements with a high lateral resolution to localize absorption centers and characterize them

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with respect to their size, density, and absorption strength. Furthermore, for a better understanding of the physical, chemical, and structural reasons for residual absorption, it is necessary to gain information about the localization in depth of the thin film system; i.e., to discriminate between thin film absorption, substrate absorption, and absorption in interface layers.

Photothermal methods<sup>11-13</sup> utilizing probe lasers for a noncontact inspection have been shown to provide many of the desired capabilities. They combine ultrahigh sensitivity with high spatial resolution and offer noninvasive tools for the inspection of optical coatings.<sup>14</sup> Because the probed volume can be controlled easily during the photothermal experiment, depth profiling is possible.<sup>15</sup> Although the lateral resolution of probe-laser-based techniques is limited to approximately 1  $\mu\text{m}$  for principal physical reasons, submicrometer depth resolution is obtainable by a proper choice of experimental parameters. Various problems of optical thin film characterization have been attacked successfully by photothermal measurements that yielded unique results. Because of the extremely high sensitivity of the advanced measurement techniques, it is possible to measure minimum absorption characteristic of optimized optical components.<sup>16-19</sup> Energetic-radiation-induced changes in the absorption behavior have been monitored.<sup>20</sup> Also, characteristic differences in the absorption for thin films prepared by different deposition techniques have been investigated.<sup>21</sup>

In recent years, photothermal imaging techniques with high spatial resolution have been developed for the nondestructive evaluation of a great variety of thin film systems.<sup>22</sup> Most of these experiments employ a modulated pump laser beam (heating beam) that is used to create thermal diffusion waves for imaging the surface as well as buried structures.<sup>23-29</sup> For the characterization of optical thin films, photothermal imaging has been used for the detection of large-scale inhomogeneities as well as localized defects in the micrometer range. It has been shown that photothermal microscopy is able to detect defects and inhomogeneities that are often not observable by conventional techniques.<sup>30-34</sup>

The purpose of this paper is the characterization of  $\text{ZrO}_2$  and  $\text{MgF}_2$  films with respect to their optical absorption properties at 514 nm wavelength with special emphasis on lateral as well as depth localization of absorption centers. Several single-layer films of  $\lambda/2$ ,  $\lambda$ , and  $2\lambda$  optical thickness deposited on BK7 glass and SQ1 quartz substrates have been investigated. It was our goal to compare results obtained for films of various film thickness and investigate the influence of the substrate material and the thin film deposition technique on defect properties.

Measurements were based on the photothermal displacement technique<sup>35</sup> and were performed on an instrument that has been specially adapted to displacement microscopy. The displacement method measures the absorption by probing the thermoelastic response of the sample surface that is heated by a focused pump laser beam. The thermoelastic deformation (or displacement) causes a small change in the reflection angle of a probe beam that is monitored by a position-sensitive device. To obtain high sensitivity and spatial resolution the pump beam was intensity modulated and only the harmonic part of the photothermal response was measured. The depth sensitivity of such measurements is controlled by a variation of the modulation frequency.

To obtain complementary information we also performed total integrated scattering (TIS) measurements on some of the films. This technique yields information about the local distribution of scattering defects but provides much lower spatial resolution. Results are compared to defect densities obtained from photothermal measurements.

## 2 Experiment

### 2.1 Photothermal Displacement Measurements

A schematic of the photothermal displacement setup is shown in Fig. 1. The cw pump light source was an  $\text{Ar}^+$ -ion laser providing up to 2-W laser light at a wavelength of 514 nm. The beam was intensity modulated harmonically by an optoacoustic modulator in the frequency range from 1 to 100 kHz and focused onto the sample surface to a spot of approximately 40  $\mu\text{m}$  diameter. The HeNe probe laser beam (focused to about the same spot diameter) was incident under 45 deg from the sample surface normal and directed to a quadrant detector after reflection. The angular deflection of the probe laser beam in the plane spanned by the incident and reflected beams as well as perpendicular to this plane was obtained by subtracting signals from opposite quadrants of the detector. All measurements presented here represent in-plane deflections. The distance between pump and probe beam focus was adjusted to a fixed value yielding maximum signal in the slopes of the surface deformation. Signals were processed by a dual-phase lock-in amplifier synchronized to the pump beam modulation. This device provided amplitude and phase (with respect to the modulation) of the displacement signal. Images were obtained by raster scans of the laser beam over the sample surface. For this purpose, the sample was mounted on an  $xy$  positioning stage allowing 20 mm of sample movement in each direction with a positioning accuracy and repeatability of  $\pm 0.1 \mu\text{m}$ . Detailed descriptions of the basic principles of photothermal displacement measurements<sup>35</sup> as well as the resolution power of general laser scanning<sup>36</sup> and photothermal techniques<sup>37</sup> can be found in the literature.

In many of our measurements it was found that the size of some defects was smaller than the pump beam diameter leading to an inhomogeneous absorption over the pump beam area. Such an absorption center leads to a strong localized heating of the thin film and can cause a distortion of the surface displacement profile. Depending on absorption

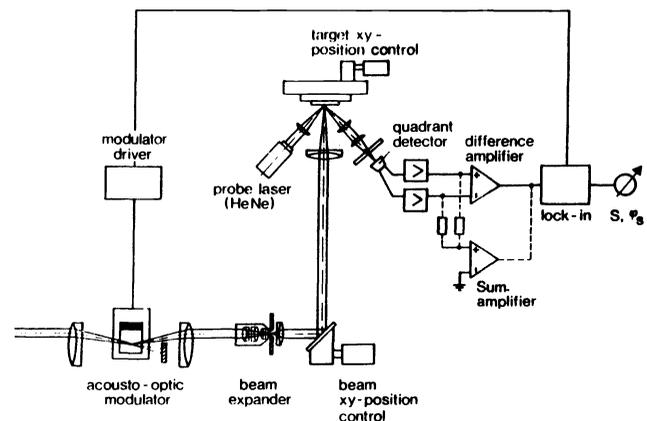
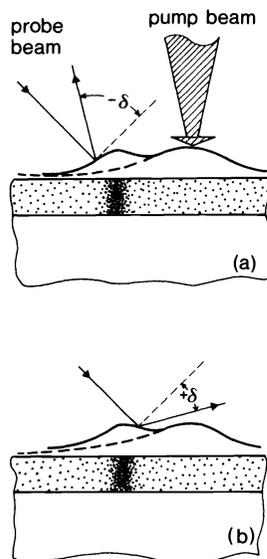


Fig. 1 Experimental setup used for photothermal displacement microscopy.

**Table 1** Synopsis of the main thin film and experimental parameters. Results for absorption contrast, diameter, and number density of inhomogeneities represent averages over various scans. Numbers in parentheses are values for the uncoated substrate. Temperatures  $T$  denote substrate temperatures during evaporation.

set #	substrate/coating		deposition technique	absorption contrast	diameter of inhomogeneities [ $\mu\text{m}$ ]	area density of inh'ties [ $\text{mm}^{-2}$ ]			
						$f=1\text{kHz}$	$f=10\text{kHz}$	$f=40\text{kHz}$	$f=100\text{kHz}$
I	BK7	$2\lambda$ $\text{ZrO}_2$	laser beam evaporation	10...115	6...50	60	24	12	-
II	BK7	$\lambda/2$ $\text{MgF}_2$ $\lambda$ $\text{MgF}_2$	resistance evaporation $T=273\text{K}$	5...100	5...110	14 (8)	10	12	8
				5...120	4...96	34 (12)			
	8...94	4...90	15 (2)						
	10...105	7...120	18 (4)						
III	SQ1	$\lambda/2$ $\text{MgF}_2$ $\lambda$ $\text{MgF}_2$ $\text{MgF}_2$ , $\lambda/2$ $\text{MgF}_2$	resistance evaporation $T=570\text{K}$	4...124	6...132	16 (5)			
				2...16	4...36	10			
				3...5	8...24	5			
	BK7	$\lambda/2$ $\text{MgF}_2$		2...6	10...28	8			

strength and thermal diffusion length the shape of the normal profile can be changed just slightly or a double-peak structure may arise, as demonstrated in Fig. 2. Such a structure leads to characteristic features in the photothermal image when the sample is scanned relative to the pump beam. Depending on the relative position between probe beam and defect-induced displacement maximum a positive or negative displacement signal is recorded. Therefore, the photothermal image of the defect consists of a double-peak pattern with zero amplitude in the center, coincident with a phase jump of 180 deg. By using the photothermal displacement technique we are able to define size and position of strongly absorbing defects with a spatial resolution far below the spot diameters of the involved laser beams. In case of structural inhomogeneities



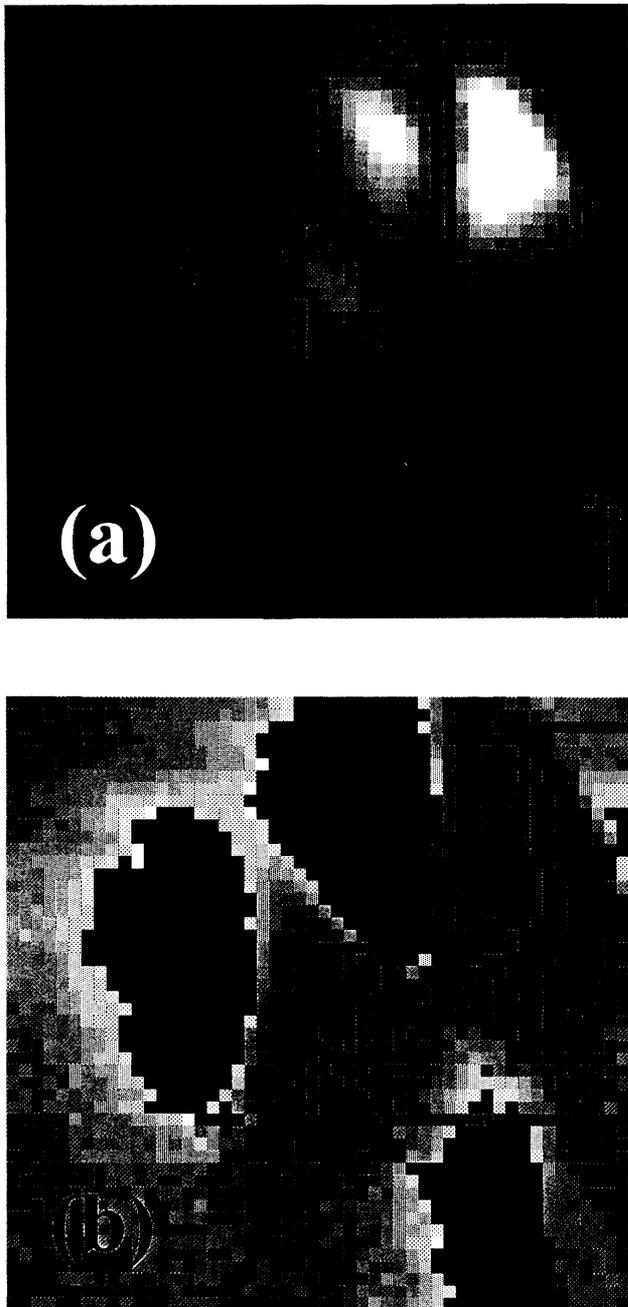
**Fig. 2** Illustration of a distorted photothermal displacement profile in the presence of a localized absorption center smaller than the undistorted deformation pattern. Depending on the relative position between probe beam and absorbing spot, a positive ( $+\delta$ ) or negative ( $-\delta$ ) angular beam deflection results (dashed line represents the unperturbed reflection without photothermal deformation). If the probe beam is coincident with the maximum of the additional peak the displacement amplitude vanishes and the phase changes by 180 deg (for a measured example see Fig. 3).

leading to thermal barriers, photothermal edge effects.<sup>38,39</sup> may result in an additional signal enhancement.

Displacement images were taken for  $\text{ZrO}_2$  and  $\text{MgF}_2$  single layers with various experimental parameters. Table 1 shows a synopsis of thin film material, thickness, and deposition technique as well as the substrates for three sets of samples (I, II, and III). For some samples scans were performed at various frequencies (listed in the table) and various spatial resolutions.

All images presented here display amplitude and phase of the photothermal displacement signal as a function of scanning position. Normalization of the gray-scale amplitude images was performed to obtain a range from zero signal (black) to maximum signal (white) in the respective region. For the phase images, the gray scale was chosen to cover the range from  $-180$  (black) to  $+180$  deg (white). A typical result taken at 1-kHz modulation frequency for a  $100 \times 100\text{-}\mu\text{m}^2$  scan on a  $\text{MgF}_2$  sample is shown in Fig. 3. We found a small but clearly measurable signal throughout the entire scanning area representing the mean homogeneous absorption of the unperturbed coating. Additionally, absorption centers at various locations can be identified clearly in amplitude as well as phase. For strong absorption centers with a diameter smaller than the pump beam radius (as present in Fig. 3), a symmetric feature with zero signal amplitude in the center line and phase jump by 180 deg is observed. This results from a local maximum in the displacement profile leading to a change in sign of the surface slope when scanning over the inhomogeneity as described in the previous section.

For a quantitative analysis of such images, we define the absorption contrast of the inhomogeneity (IH) as the ratio of the photothermal amplitude in the center of the IH to the average amplitude in the surroundings of the IH; values were extracted from high resolution scans and are compiled in Table 1. Furthermore, in Table 1 values for number density and diameter of the inhomogeneities are given. The area density of inhomogeneities varied from 5 to 34 per squared millimeter with a mean of 14 per squared millimeter over all sample sets. Considering that the bare substrate itself carries absorbing defects (see Table 1) this is well in accordance with the results obtained by Staggs et al.<sup>3</sup> in multilayer coatings. It can be assumed that the photothermal method yields larger numbers of defects than AFM, because the latter technique is solely sensitive to structural inhomogeneities such



**Fig. 3** Photothermal displacement image ( $100 \times 100 \mu\text{m}^2$ ) of a  $\text{MgF}_2$  film on BK7 at 1-kHz modulation frequency (a) amplitude and (b) phase. The well-defined phase over the entire picture indicates a nonzero absorption signal. The phase jump of 180 deg present in the main feature results from a small absorbing spot.

as nodule defects, whereas the former is also able to detect absorption centers not accompanied by morphological irregularities. This assumption is supported by the observation of large-diameter inhomogeneities present in most of our scans. According to a general model<sup>4</sup> nodule defects with diameters of more than  $100 \mu\text{m}$  are very unlikely in a single-layer coating and probably result from other types of defects. To clarify this point a comparative study of defect morphology by AFM and defect absorption profile is necessary.

## 2.2 Total Integrated Scattering Measurements

Total integrated scattering (TIS) measurements were performed with the sample placed in a Coblenz sphere as illustrated in Fig. 4. The beam of the primary light source (HeNe laser) was directed onto the sample (spot size 1.6 mm) with an incidence angle close to 0 deg to guide the specular reflection through the same small hole in the Coblenz sphere as the incoming beam. Collection of the scattered light by the sphere covered the range of 2 to 84 deg from the sample normal. The total scattered intensity  $I_{sc}$  was monitored by a photomultiplier. To enhance sensitivity the primary laser beam was intensity modulated by a mechanical chopper and the photomultiplier signal processed by a lock-in amplifier. By normalization, we calculated the TIS signal that we define as

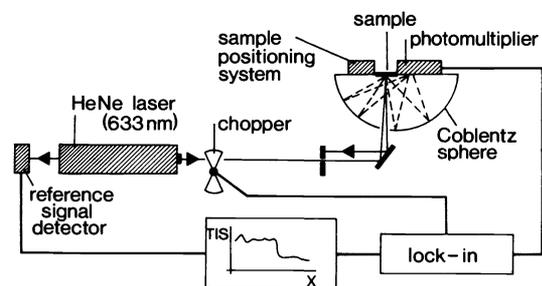
$$\text{TIS} = \frac{I_{sc}}{I_0},$$

where  $I_0$  is the intensity of the incoming HeNe laser beam. Laterally resolved scattering results were obtained by mounting the sample on a positioning stage allowing one-dimensional scanning. To investigate the dependence of scattered light on film thickness,  $\text{MgF}_2$  films with two different thicknesses on BK7 substrates were overcoated with a 70-nm Al overlayer prior to the TIS measurements. In that way, thickness dependent scattering interference effects were avoided and a value for the evolutionary exponent  $\kappa$  could be obtained.<sup>40,41</sup>

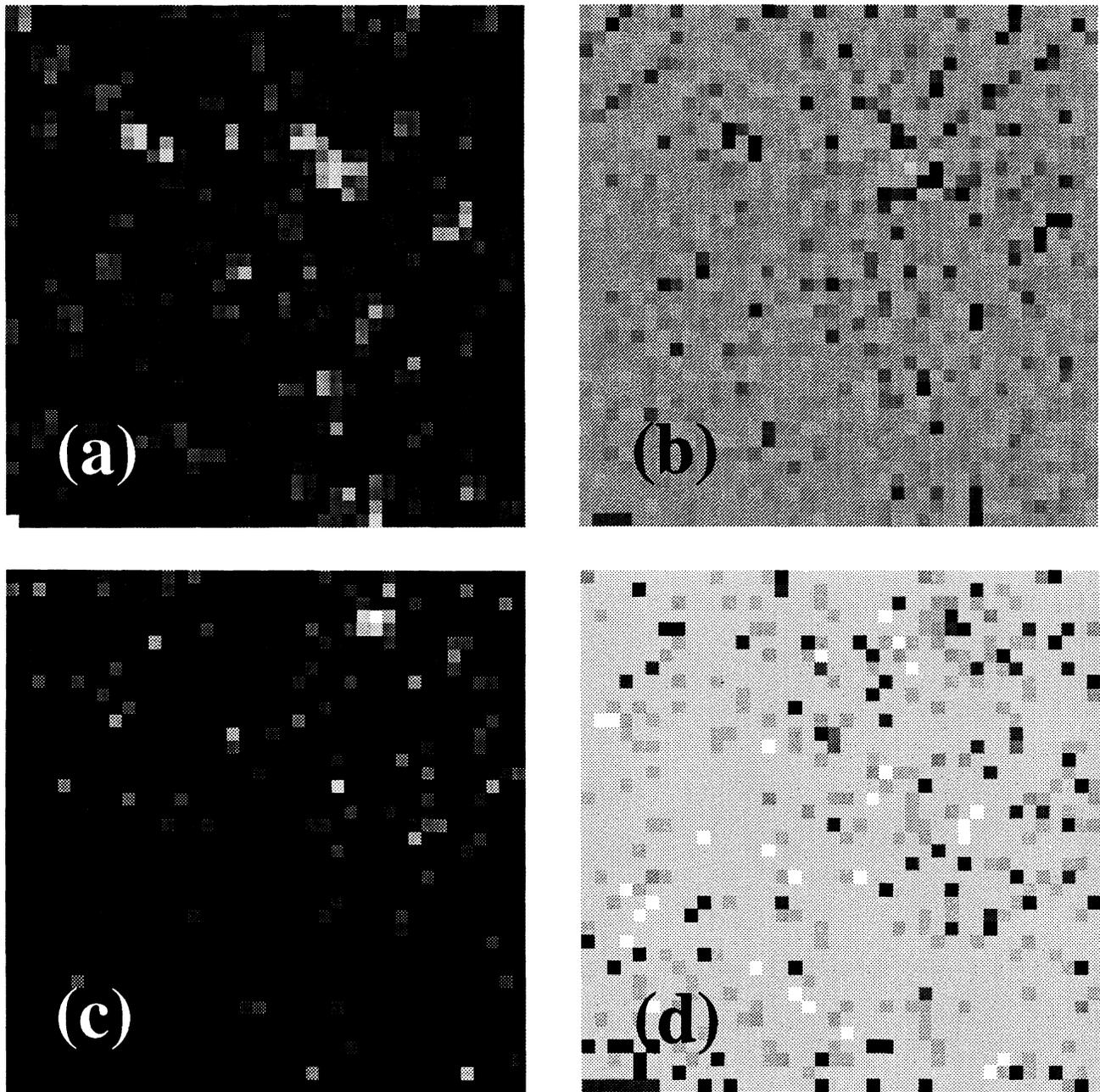
## 3 Results

### 3.1 Displacement Images of $\text{ZrO}_2$

Photothermal images of large-area scans on sample I (see Table 1) at 1 and 10 kHz are shown in Figs. 5(a), 5(b), 5(c), and 5(d), respectively. These images give a general overview of the absorption behavior and distribution but cannot be used for an exact defect density determination because of insufficient lateral resolution. However, from the amplitude representations, it is evident that the area density of defects is much higher for a modulation frequency of 1 kHz than for 10 kHz. These findings are also supported by the analysis of images taken at 1 and 40 kHz with better spatial resolution, as displayed in Fig. 6. Here, pixel size is small enough to resolve individual defects and to determine absorption contrast as well as diameter and area density of the inhomogeneities. From the values in Table 1, we find a systematic



**Fig. 4** Experimental setup used for TIS measurements.

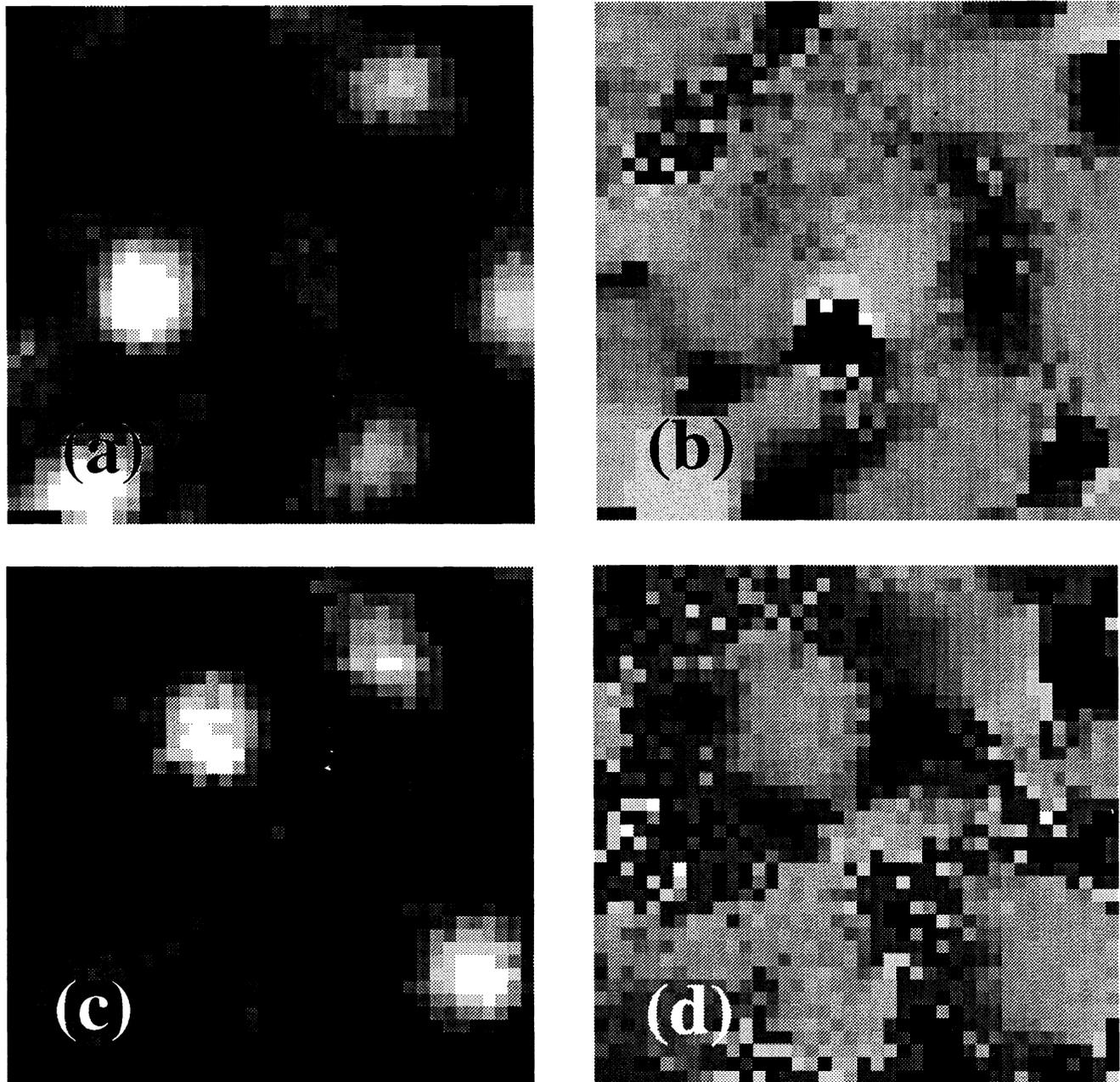


**Fig. 5** Displacement amplitude (a) and (c) and phase (b) and (d) images on  $\text{ZrO}_2$  taken at (a) and (b) 1 kHz and (c) and (d) 10 kHz. Scanned area (a) and (b),  $1 \times 1 \text{ mm}^2$ , and (c) and (d),  $2 \times 2 \text{ mm}^2$ .

decrease of area density of IHs with increasing frequency. Note that not only these parameters change with frequency but also the apparent phase contrast is higher at higher modulation frequency.

For an interpretation of these results, we recall that the thermal length, i.e., the depth range where the thin film system is probed, depends on modulation frequency and thermal parameters of the sample.<sup>42</sup> For a  $1\text{-}\mu\text{m}$   $\text{ZrO}_2$  film, thermal conductivity has been determined recently and a very low value of  $5 \times 10^{-2} \text{ Wm}^{-1} \text{ K}^{-1}$  was found.<sup>43</sup> The resulting thermal length is about half of the film thickness when operating at 10 kHz but larger for a modulation frequency of 1 kHz. Assuming that the photothermal signal is generated

at absorption sites in various depths with different densities for thin film, interface, and substrate it is expected that scanning the thermal length across characteristic dimensions of the thin film system (i.e., thin film thickness) will result in measurable signal changes. Therefore, measured amplitude, number density, and observed lateral dimension of defects as well as the phase of the photothermal signal are expected to be a function of the modulation frequency. Note, however, that exact predictions for the photothermal displacement response of thin film systems are not yet available and depth profiling sensitivity is generally poor.<sup>44</sup> Large effects are only expected in special cases with a very specific configuration of thin film and substrate thermoelastic parameters.



**Fig. 6** Displacement amplitude (a) and (c) and phase (b) and (d) images on  $ZrO_2$  taken at (a) and (b) 1 kHz, and (c) and (d) 40 kHz. Scanned area:  $200 \times 200 \mu m^2$ .

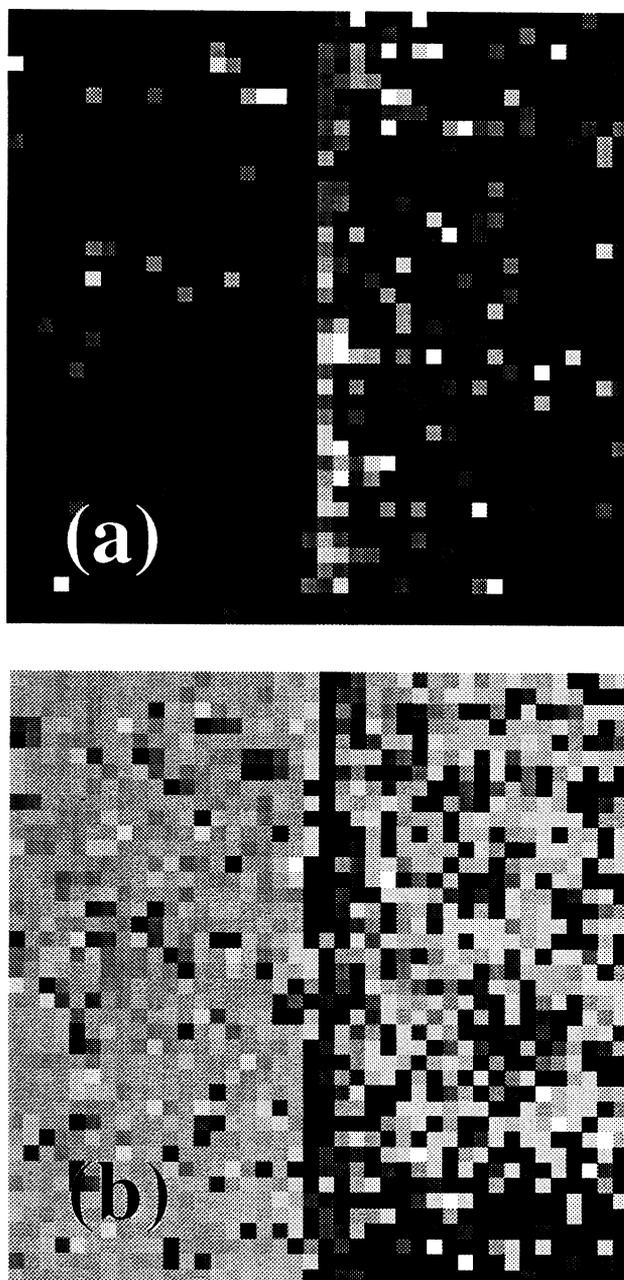
### 3.2 Displacement Images of $MgF_2$

Most measurements were performed on  $MgF_2$  thin films. A typical example is shown in Fig. 7 with images for a  $MgF_2$  thin film of  $\lambda$  optical thickness on BK7. During preparation of this sample one half of the substrate was covered and, therefore, photothermal signals from both the thin film system and the bare substrate material could be studied on one sample. We found that the mean signal amplitude on both sides of the boundary of coverage was about equal, indicating that the evaporated thin film did not increase the mean absorption significantly. However, the density and absorption strength

of defect centers is considerably larger on the coated side than on the bare substrate material. Furthermore, we observed a strong accumulation of defects at the boundary line of the coated area. We believe that this provides evidence for a defect diffusion process during thin film evaporation; i.e., in addition to defects already present on the substrate material the evaporation process introduces new impurities that diffuse on the substrate surface and are trapped at the boundary.

### 3.3 TIS Measurements on $MgF_2$ Films

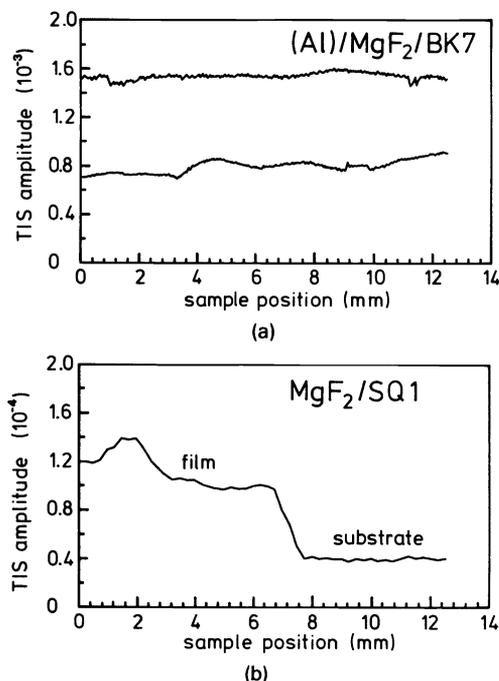
For a more detailed investigation of the substrate influence,



**Fig. 7** Amplitude (a) and phase (b) images of the displacement signal for a  $\lambda$   $\text{MgF}_2$  film on a BK7 substrate. The right side of the substrate was coated, whereas on the left side the bare substrate was measured. Note the accumulation of defects at the boundary. Scanned area:  $1 \times 1 \text{ mm}^2$ .

TIS measurements were performed on  $\text{MgF}_2$  films of two different thicknesses on BK7 substrates and one film on quartz; results are shown in Figs. 8(a) and 8(b).  $\text{MgF}_2$  films on BK7 substrates [Fig. 8(a)] exhibit high and thickness-dependent TIS intensity of the order  $10^{-3}$ . From the measurements we calculate a value of  $\kappa = 0.4$ ; i.e., the mean dimension of structural features (columns)  $D$  of the  $\text{MgF}_2$  films grows as  $D \sim d^{0.4}$ , where  $d$  is the film thickness.

Variations of the scattering amplitude with the position on the sample are small, especially in the case of the thick sample. This is in accordance with spatially resolved photo-



**Fig. 8** Total integrated scattering measurements on  $\text{MgF}_2$  films on BK7 (a) and SQ1 (b) substrates. (a) Sample thickness of films on glass substrates was 690 nm (upper curve) and 410 nm (lower curve). (b) One half of the quartz substrate was covered during deposition of the  $\text{MgF}_2$  film of  $\lambda$  optical thickness ( $\lambda = 514 \text{ nm}$ ).

thermal absorption measurements on these samples that also revealed a very smooth absorption structure. The average TIS values of  $\text{MgF}_2/\text{BK7}$  layers of  $\lambda$  thickness without Al overcoating was  $3 \times 10^{-5}$  and again displayed only small local variations over the sample surface.

#### 4 Discussion

From the analysis of high-resolution scanning data compiled in Table 1 three conclusions about defect properties of the  $\text{MgF}_2$  layers can be drawn. First, note that measurements performed on the  $\lambda/2$  sample from set II do not exhibit a significant frequency dependence of the apparent defect densities. This probably results from the fact that the  $\text{MgF}_2$  layer has a much larger thermal conductivity<sup>45</sup> than  $\text{ZrO}_2$ . Therefore, the  $\text{MgF}_2$  layer can be regarded as thermally thin for all the frequencies we used and depth profiling effects are not to be expected. Furthermore, the large coefficient of thermal expansion of  $\text{MgF}_2$  and its influence on photothermal signal generation in layered systems should be noted. Second, from a comparison of data from sets II and III it follows that defect properties depend strongly on the fabrication process. Defect densities found for samples evaporated at 273 K substrate temperature are larger by a factor of 2 compared to those for films evaporated at 570 K. Also, mean diameter and absorption contrast change drastically with substrate temperature. As a third finding, we mention slight differences in absorption properties for  $\lambda/2$  samples from set III evaporated on BK7 and SQ1 substrates, respectively. Small but significant reductions of defect densities, diameter, and absorption contrast are observed between the films on the two different substrates. The quality of the film on the BK7 substrate is better than for the SQ1 substrate. We believe that this effect

results from the tendency of SQ1 to electric charging, a process leading to an accumulation of impurities at the substrate surface, as observed in Fig. 7.

TIS signals of the half coated MgF<sub>2</sub>/SQ1 system were about one order of magnitude larger than those of the films on BK7 substrates. In Fig. 8(b), the pronounced step in TIS amplitude indicates the boundary between coated and uncoated regions. No defect accumulation is observed at the boundary. This demonstrates that photothermal and TIS measurements yield truly complementary results, i.e., the first method is solely sensitive to absorbing defects, whereas the latter is a probe for scattering only. In the MgF<sub>2</sub> covered region of the sample, a strong variation of TIS amplitude with sample position is observed at one position, indicating a region of increased scattering defect density.

## 5 Summary

ZrO<sub>2</sub> and MgF<sub>2</sub> single-layer films on glass and quartz substrates have been investigated by photothermal displacement microscopy and total integrated scattering measurements. It was demonstrated that photothermal microscopy is able to reveal images of localized absorption centers with several micrometers lateral extension. From the photothermal images, values for mean defect densities, mean diameter, and absorption contrast could be derived.

It was found for a ZrO<sub>2</sub> film that the measured defect density is a function of modulation frequency, indicating that defects in different depths of the thin film system can be probed. Great differences in defect properties of MgF<sub>2</sub> thin films for samples prepared with different process parameters were observed. Furthermore, a slight dependence on substrate material could be extracted from the data, whereas results were independent of thin film thickness. Complementary information was obtained by TIS measurements.

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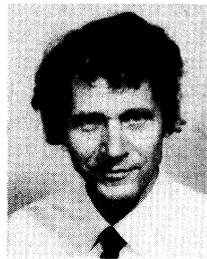
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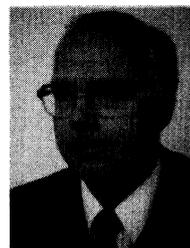
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