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Ripple Structures and Enhanced Absorption Associated With Ordered Surface Defects

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Laser-induced ripple structures on material surfaces have been observed by a number of workers for various materials including metals, semiconductors and dielectrics. A model has been proposed [1] which correctly accounts for the spacing and polarization dependence of the ripples, and the association of the ripples with material defects. In this model the ripples are associated with the interference of the incident field with the nonradiative field induced by the interaction of the incident field with the material defects. The association of these features with defects is often difficult to see since real optical surfaces have many defects having a range of sizes and orientations. In this paper experimental results which unambiguously show the association of these features with defects are presented. Quantitative measurements of laser-induced damage thresholds and photoacoustic absorption measurements show a reduction of damage thresholds and increased optical absorption for surfaces with controlled, linear defects aligned orthogonal to the incident laser field (as predicted in ref. 1).

Key Words: Laser-generated ripples; surface damage; surface defects; NaCl; ZnS; ZnSe; CdTe; 1.06 μm; 10.6 μm.

1. Introduction

Laser-generated ripple patterns (LGRP) have been observed in laser-induced damage (LID) to surfaces of various wide bandgaps dielectrics [1-14], semiconductors [14-26], and metals [1,2,14,25,26,28-36]. These features have also been observed in dielectric films used as anti-reflection coatings [6-9]. Careful examination of micrographs in a number of earlier LID papers reveal LGRP on surfaces which were initially unnoticed or were wrongly interpreted as features which were caused by multiple reflections within the sample [37-41]. Recent work has shown these features to be common artifacts of laser annealing experiments, while other experiments have revealed LGRP associated with laser-assisted thin film deposition [36]. The observation of LGRP over such a large range of wavelengths (0.17 to 10.6 μm) and such a wide range of materials has prompted Van Driel to speculate that LGRP are a universal phenomenon common to all laser-surface interactions [42].

Most of the LID experiments referenced above were conducted using optically polished surfaces. Such surfaces, particularly those on exotic materials such as the alkali-halides, contain a high density of defects, e.g., scratches, digs and residual polishing material. LGRP are found to be directly associated with these pre-existing surface defects. Figure 1 shows LGRP running parallel to scratches which are orthogonal to the impressed laser electric field, whereas no LGRP are produced by the same type defect parallel to the field. Similar features have been observed for point defects and grain boundaries in polycrystalline materials.

The observations above lead to a model by Temple and Soileau [1,2] for the formation of LGRP that assumes that these features are initiated by the interaction of the impressed field with pre-existing surface defects. A similar, much more rigorous, model has been proposed by Sipe et al [43]. In that work, the authors show that ripple formation may be initiated by the components of the surface sub-microscopic roughness which are oriented for optimum coupling with the impressed field. Other models [12,36,44] have been recently proposed and these all have the common feature that LGRP result from some sort of scattered wave launched by surface roughness (i.e., submicroscopic surface defects) which then constructively interferes with the impressed field to produce ripples.

The recent models have the common feature that they predict that LGRP are initiated by surface roughness [12,36,43,44]. The model proposed by Temple and Soileau [1,2] predicts that defects of specific sizes and orientation with respect to the incident field should have maximum efficiency for creating LGRP (the model by Sipe et al [43] has a similar provision which they refer to as the "efficacy factor"). Reference 1 predicts that LGRP are easiest to form for scratches of width equal to the laser wavelength and oriented orthogonal to the electric field of a linearly polarized beam. The effects of scratch orientation on LGRP generation on dielectric surfaces have been reported in measurements of the LID thresholds of diamond-turned optical surfaces [10] and more recently in specially prepared Si surfaces [23].

In this paper we report the results of measurements of the thresholds for LGRP formation on NaCl surfaces. We attempted to make the defects nearly "resonant" with the impressed laser field by grinding the NaCl surface with particles of average diameter approximately equal to the laser wavelength (10.6 μm). In addition, we made the observation of LGRP in ZnS, ZnSe, CdTe and other materials.

2. Experimental

A single crystal NaCl specimen was first etched to remove most residual, randomly oriented scratches, due to polishing by the crystal vendor. The sample was then ground on the edge of a polishing wheel using 10 μm alumina grit so as to produce a high density of parallel scratches. A linearly polarized CO_2 TEA laser operated in the TEMoo spatial mode at 10.6 μm was used to irradiate this specially prepared sample. The laser was gain-switched with a partially mode locked temporal profile. The laser was operated without N_2, and the temporal width of the gain-switched envelope was approximately 100 nsec (FWHM). The laser was focused onto the exit surface of the sample. The fluence was varied using a set of Brewster angle polarizers arranged so as to keep the orientation of the laser electric field vector constant at the sample surface. The threshold fluence for the onset of LGRP formation was then measured for the cases where the surface scratches were oriented parallel to and orthogonal to the laser electric field.

This same laser system was used to produce LGRP on the exit surfaces of optically polished ZnSe and ZnS. In addition, a Nd:YAG laser was used to produce LGRP on various materials. The Nd:YAG laser was linearly polarized and operated at 1.06 μm in the TEMoo spatial mode (this laser is more completely described in ref. 45).

The CO_2 laser system described above was also used as the source for orientation dependent absorption measurement for an aluminum diffraction grating. The grating spacing was 9 μm. The absorbed energy was monitored using a photoacoustic technique. The laser-induced acoustic signal was detected by a piezoelectric tranducer pressure contacted with vacuum grease to the rear surface of the metal grating. The absorption, as determined by the piezoelectric peak voltage output, was measured for the cases where the grating grooves were parallel to and orthogonal to the laser electric field.

3. Results and Discussion

Figure 2 is a Nomarski micrograph of LGRP on the specially prepared NaCl surface. The micrograph on the right corresponds to the situation for which the parallel scratches are orthogonal to the incident field vector. Note that as a scratch enters into the high field region it seems to "grow" until its width is approximately equal to the wavelength of the light in the material. A well coordinated ripple pattern results since adjacent ripples are optimumly spaced for maximizing the induced surface field. The micrograph on the right is a picture of a damage sight for the situation where the scratches are parallel to the incident field. The only ripples seen in this micrograph are associated with a scratch (probably left over from the original polishing) which is nearly normal to the impressed field.

We measured the threshold fluence for the onset of LGRP with the laser field parallel to and orthogonal to the parallel scratches. The results of these measurements are shown in figure 3. Note that the threshold for ripple formation is 40% higher for the case where the scratches are oriented parallel to the impressed electric field.

The above result is consistent with the model presented in reference 1 and can be understood in terms of the well known phenomenon of the orientation dependence of absorption of a diffraction grating [46]. Figure 3 is a plot of the 10.6 μm absorption of an aluminum grating with 9 μm grating spacing. The absorption was measured for the cases where the grooves are parallel to and normal to the laser electric field for linearly polarized light. As can be seen in figure 4, the absorption is 4.6 times larger for the case where the grating lines are ortogonal to the impressed field compared to the case where the grating lines are parallel to the incident laser field. So, for this specimen the surface absorption is 4.6 times greater for the case where the defects (i.e., the grating lines) are normal to the incident field. By comparing the results shown in figure 4 with those shown in figure 3 we see that the threshold for initiating LGRP is lowest for the same orientation of the surface defects (scratches) which lead to maximum surface absorption. As the ripples form, coupling of light into the surface increases, and this in turn enhances the formation of more ripples. In fact, an exponential growth of LGRP has been observed [12].

LGRP observed in dielectrics, semiconductors, and metals are very similar in that they run normal to the impressed field and are associated with surface defects. They differ in one important way: the ripple spacing is the free space wavelength (λo) for semiconductors and metals, whereas their spacing is $\lambda o/n$, (n is the index of refraction), in the wide bandgap dielectrics. Table 1 summarizes the materials for which LGRP have been observed in our lab and the observed ripple pattern spacings. One may speculate about the reason for the difference. One possibility is that the semiconductors undergo a semiconductor to metal phase transition prior to ripple formation. The model given in reference 43 predicts that a transition from λo is $\lambda o/n$ spacing occurs for index of refraction between 4 and 1.5. We attempted to observe the occurance of LGRP with both spacings by examining materials of intermediate index. CdTe and ZnSe were examined at 1.06 μm, and ZnSe and ZnS were examined at 10.6 μm. In addition ZnSe has been studied at 3.8 μm. In all cases the LGRP observed had spacings of λo for these materials.

4. Summary

We have studied LGRP on surfaces with oriented defects (parallel scratches). The threshold for initiation of LGRP was 40% lower for the case where the defects are normal to the impressed laser electric field. Scratches normal to the field produce well coordinated ripple patterns which are clearly associated with the scratches. Photoacoustic measurements of absorption at 10.6 μm of an aluminum grating with 9 μm spacing show that surface absorption is 4.6 times larger for the grating lines normal to the field vector. Thus, we see that LGRP formation is easiest for defect orientation for which surface absorption is maximum.

Ripple patterns were produced on surfaces of CdTe and ZnSe at 1.06 μm and on surfaces of ZnSe and ZnS at 10.6 μm. In all cases the ripple spacing was λo, the free space wavelength. Ripples observed on metals and semiconductors all have a spacing of λo, whereas those observed on wide bandgap dielectrics are spaced at $\lambda o/n$.

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5. References

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(NaCl)

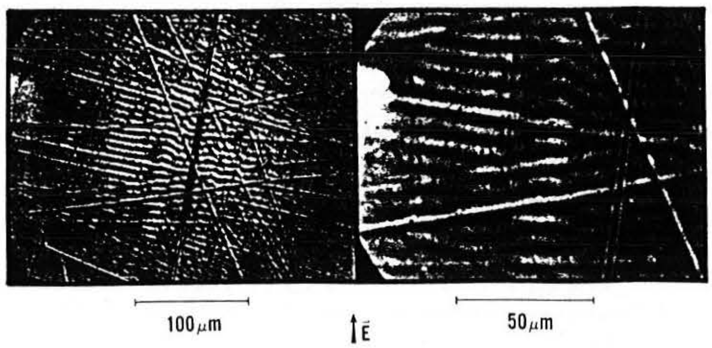

Figure 1. Laser Generated Ripple Patterns (LGRP) in NaCl. The above patterns were produced by normally incident 10.6 μm laser light on the exit surface of NaCl. The laser light was linearly polarized in the direction shown by $\vec{E}$. The higher power photo on the right illustrates that the ripples are associated with defects normal to $\vec{E}$ whereas scratches parallel to $\vec{E}$ have no ripples associated with them.

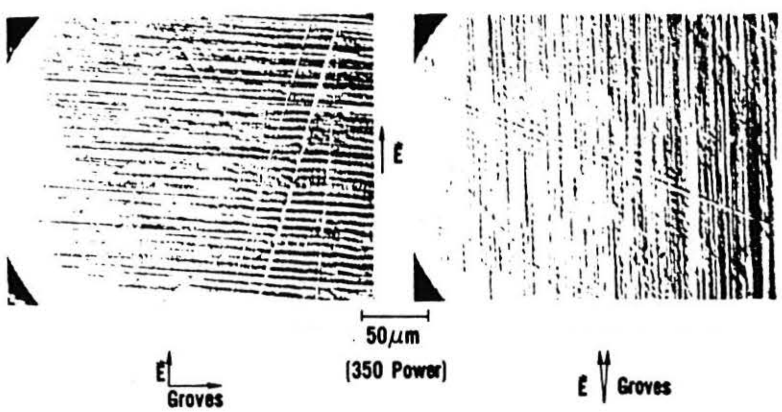

Figure 2. LGRP's on the exit surface of a NaCl specimen. The laser wavelength was 10.6 μm. The laser was linearally polarized in the direction indicated by the arrow ($\vec{E}$). Each site was irradiated only once.

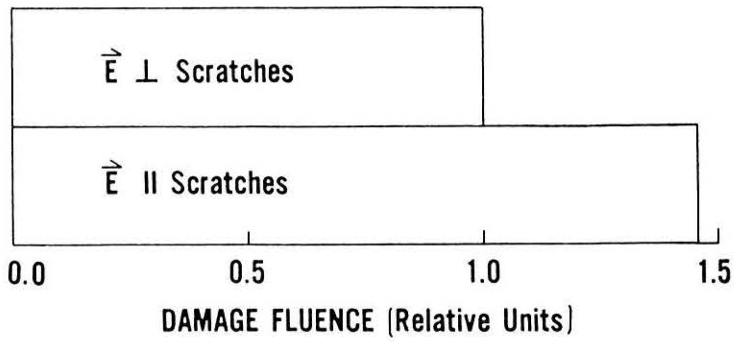

Figure 3. Threshold for Formation of LGRP's. The above is a plot (in relative units) of the fluence required to produce the LGRP for the cases where the linearly polarized electric field is parallel to and normal to the surface scratches. Measurements were made on a NaCl sample (exit surface) using 10.6 μm radiation.

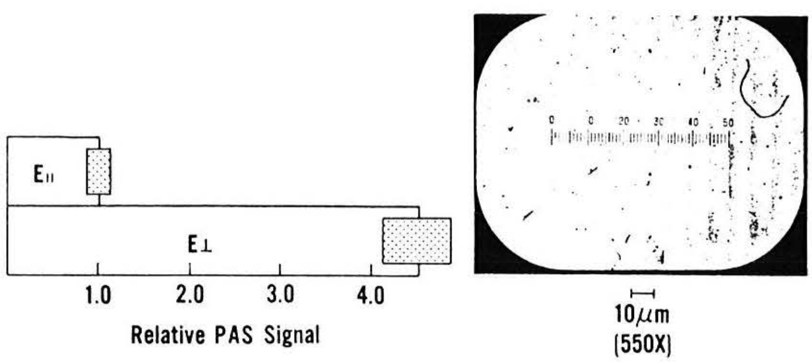

Figure 4. Grating Absorption. The bar graph on the right is the PAS signal (in relative sites) for normally incident, linearly polarized, 10.6 μm laser radiation. The micrograph on the right shows the surface of the Al grating.

Material	1.06μ	2.7μ	3.8μ	10.6μ
$KC\ell$	λ_o/n			λ_o/n
$NaC\ell$	λ_o/n		λ_o/n	λ_o/n
KBr	λ_o/n			λ_o/n
SiO_2 (fused)	λ_o/n			
BaF_2		λ_o/n	λ_o/n	
SrF_2		λ_o/n		
MgF_2			λ_o/n	
Si	λ_o			
CdTe	λ_o			
ZnSe	λ_o			λ_o
ZnS				λ_o
CdS	λ_o			
$Cd(S_{.25}Se_{.75})$	λ_o			
ZnTe	λ_o			
$A\ell$	λ_o			λ_o
Cu				λ_o
Ag				λ_o

Table 1. The above is a listing of the materials, wavelength, and LGRP spacing observed in this work. Additional materials have been studied by other workers (see the references listed in the introduction).