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Aligned, coexisting liquid and solid regions in laser-annealed Si

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Aligned liquid and solid stripes are observed under continuous-wave laser illumination of Si films. For normally incident light, the stripes have a periodicity equal to the laser wavelength and are aligned perpendicular to the laser polarization. The dependence of the stripe spacing versus incident angle is also probed. Examination of the surface after cooling shows the presence of surface ripples with the same wavelength. Thus it is proposed that the liquid and solid Si stripes are the precursors of the ripples. It is suggested that aligned liquid-solid regions are obtainable for certain conditions for pulsed laser annealing of silicon.

An unusual effect that has been observed from both continuous-wave (cw) and pulsed laser annealing of semiconductors is the presence of periodic surface structures.[1-4] For normally incident linearly polarized laser radiation, the surface structures are linear, parallel ripples with the peaks separated by a distance equal to the wavelength of the annealing radiation. Several models based on standing surface electromagnetic waves have been proposed to account for the surface ripples.[2,3,5] It is difficult, however, to explain how surface topography can be maintained in molten Si after the radiation is removed.[6] In this study *in situ* visualization of the cw-laser-annealing process is utilized to identify the precursors of the surface ripples.

The samples used in this study consisted of 0.5 to 2-μm-thick polycrystalline Si films deposited on 3-in. quartz substrates 0.4 mm thick. To achieve *in situ* visualization, 10.6-μm radiation from a CO_2 laser was focused onto the polycrystalline Si film. The laser was focused to circular or "elliptical" spots using a spherical or cylindrical lens. Care was taken to minimize wave-front distortion. A microscope (10 to 20 $\times$) objective was positioned behind the substrate, and the laser-annealed spot was imaged on a video camera. The image was obtained by reflected light illumination or blackbody emission. The display was monitored in real time and recorded on video tape. The substrate was mounted on a microprocessor-controlled, motorized x-y translation stage which also had tilting capability of up to 40° from normal.

Consider first the experiments for normal incidence of the 10-μm radiation. When the laser shutter is opened, the radiation is absorbed in both the quartz substrate and the Si film, but as the sample is heated, the absorption occurs totally in the Si. The video image of the blackbody radiation shows a uniform bright region in the center of the heated spot. After a few seconds dark circular regions of approximately 3 μm diam nucleate at random spots. The spots are liquid regions which have a lower emissivity due to the increase in reflectivity upon melting. These regions move and coalesce into larger regions which form into a stable pattern of aligned liquid and solid stripes. An image of the pattern is displayed in Fig. 1(a). The distance between the center of the liquid regions is ~ 10 μm, and the orientation is perpendicular to the incident polarization. While the stripe pattern forms slowly under constant illumination, the pattern can be stimulated to form by alternately blocking and opening the beam several times. The pattern after forming was stable for periods of minutes. Furthermore, when the sample was translated at a rate less than ~ 20 μm/sec, the stripes extended themselves in-phase and "pinned" to the sample. When the beam is blocked and the sample cooled, the remaining surface topography is clearly evident in a reflected light image of the sample. An image of the topography remaining is shown in Fig. 1(b). It is clear that the surface ripples correspond to the liquid-solid coexistence regions. In contrast, when a distorted wave front is used, the liquid and solid regions remain random, and no aligned surface

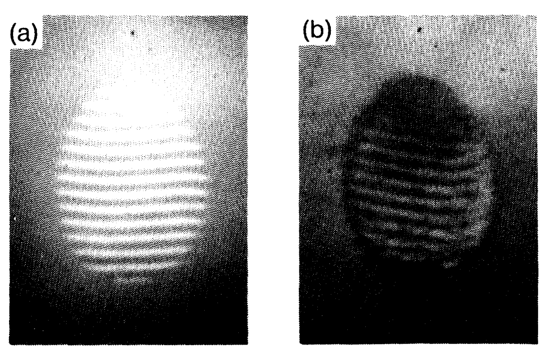

FIG. 1. (a) Magnified video image of a Si film on a glass substrate which is illuminated with ~ 3 W of 10.6-μm radiation from a CO_2 laser. (b) Resulting surface topography after cooling. In (a) the bright regions are liquid, and the spacing is ~ 10 μm.

ripple pattern is observable after cooling. The pattern is similar to the lamellar liquid or solid regions which have been previously observed.[7,8]

To determine whether the SiO_2 substrate plays a role in the interactions, the Si film was etched away and the sample was examined under Nomarski microscopy. The laser-annealed region showed random surface cracks but no evidence of ripple structures. The real time visualization showed that the surface cracks formed a few seconds after the annealing light was blocked. While coherent interactions yielding surface structures have been previously observed on SiO_2, the free-carrier absorption of hot Si precludes this interaction in the results described here.[9]

An intriguing result from pulse annealing of semiconductors is that the ripple wavelength is dependent on the annealing angle.[2,3] We have repeated the cw annealings at several angles. It was generally more difficult to observe the aligned liquid and solid regions by the *in situ* visualization, but in all cases patterns were observed with apparent spacings similar to that for normal incidence. To determine whether surface topography changes actually occurred, samples were examined by laser light diffraction.[4] Here ~5 mW of 647-nm radiation from a krypton-ion laser was focused to ~200-μm spot. The diffraction patterns were examined on screens placed ~40 cm in front or behind the sample. All the samples showed lines corresponding to diffraction from linear structures aligned in the same direction as the liquid-solid regions. From these measurements the ripple spacings were determined, and the results are shown in Fig. 2.

The major aspects of the results described here are

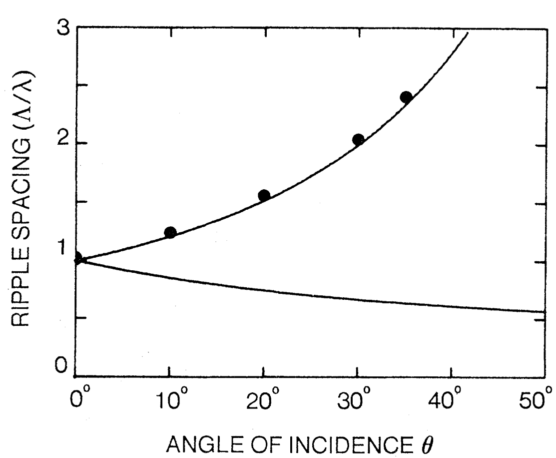

FIG. 2. Normalized ripple spacing (Λ/λ) as a function of angle of incidence (θ) for cw laser annealing of Si films. The angle θ is the deviation from normal incidence. The filled circles are the data while the solid lines represent the wavelength of standing surface waves. The ripple spacing was determined by laser light diffraction from the residual surface topography.

the coexistence of liquid and solid regions and the alignment into stripe patterns. It has previously been shown that the increase of reflectivity upon melting leads to a range of incident laser power where the solid and liquid phases coexist.[8] Simple electromagnetic and thermal considerations indicate that ~3-μm-diam liquid (solid) regions will exist in a solid (liquid) "sea" independent of light wavelength. There must be, however, a different mechanism which leads to alignment.

It has been shown that the interference between a surface and normally incident wave results in a standing-wave pattern at the surface with the same wavelength.[10] Furthermore, a surface grating structure with the same wavelength will diffract the normally incident radiation along the surface. Thus, if a surface fluctuation occurs which diffracts a portion of the incident light along the surface, the standing-wave pattern can form. If the fluctuation can couple to the standing wave, then positive feedback can occur and the fluctuation grows.[10]

In the case of laser annealing of Si, there are two aspects which will contribute to the diffraction of the incident light along the surface. These are the volume contraction due to melting and the reflectivity difference between the solid and liquid. It has been shown that the volume (height) change will result in a positive feedback situation.[10] Furthermore, considerations similar to those presented by Guosheng, Fauchet, and Siegman[10] indicate that the reflectivity change will also couple with positive feedback; thus both effects will contribute to organizing the solid and liquid regions into aligned patterns.

The change in ripple spacing versus incident angle can also be explained by the same interference properties. For waves traveling along the surface with k in the plane of the incident and reflected laser-annealing beams, the solution becomes[3]

$$\Lambda = \lambda/(1 \pm \sin\theta) \quad , \tag{1}$$

where λ is the incident laser wavelength, Λ is the wavelength of the standing wave, and θ is the angular deviation from normal incidence. The two solutions are due to traveling waves in opposite directions. The two solutions are plotted in Fig. 2. As is evident the data fit to the upper branch. Furthermore, because the visualization angle remained fixed, the observed liquid and solid regions corresponding to the upper branch should vary only slightly. The reason the longer wavelength solution is preferred here may be due to the fact that solid or liquid regions less than 3 μm diam are unstable.

The actual formation of the surface topography is then due to evaporation and/or to mass flow driven by surface tension. Microscopy of the annealed samples shows a meniscuslike surface on the region between the peaks of the ripples. In addition, in some cases the polycrystalline grain topography was still evident in the ripple "peak" regions, indicating that these regions had never melted.

The similarity of the properties of surface topography from cw and pulse laser annealing leads one to suggest that similar effects may be occurring. The obvious differences between the two procedures are that the fields are much stronger and a steady-state condition is never reached in the pulse annealing configuration. Clearly, the enhanced fields will make these effects more likely since the coupling to the field for aligned liquid and solid regions should exhibit the same positive feedback for pulse annealing. Additionally, since the energy is deposited in a short time, the large latent heat of melting of Si will be an additional factor in stabilizing the coexistence of the liquid and solid regions. Thus it seems likely that pulse laser-annealing configurations are obtainable to produce aligned, coexisting liquid and solid regions. We suggest that the presence of surface ripples may be an indication of this condition.

In conclusion, we have observed aligned, coexisting liquid and solid regions in cw laser-illuminated Si. These regions exhibit properties similar to the surface ripples observed after annealing, and it is proposed that the liquid and solid regions are the precursors of the ripple structures. Furthermore, it seems likely that conditions are obtainable in pulse laser experiments that also achieve aligned, liquid and solid regions, and as such, optical experiments may be affected by this condition.

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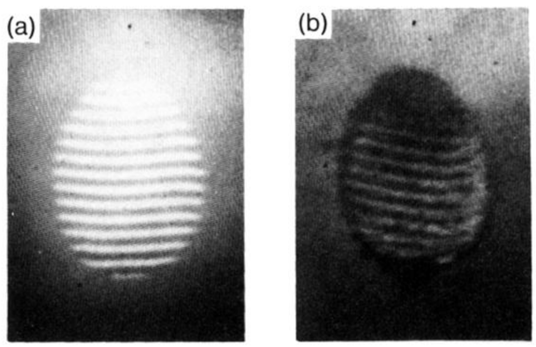

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