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# Excimer-laser-induced micropatterning of silicon dioxide on silicon substrates

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**Abstract.** Highly resolved micropatterns induced on $SiO_2$-coated Si sample surfaces have been investigated using a KrF excimer laser ($\lambda$: 248 nm and $\tau$: 23 ns). Uniform micropatterns were observed to form in the oxide layer after laser-induced melting of interfaces. The pattern size can be controlled either by the laser parameters or even by the oxide layer thickness. SEM analysis identified that the micropatterns were virtually initiated at the molten interface and the oxide layer followed the interface patterning to change its profile. Simulation of laser interaction with double-layered structures indicated that the oxide layer could melt or be ablated due to interface superheating when it was deposited on a highly absorbing Si substrate. IR analysis has demonstrated that the structural properties of the $SiO_2$ layer undergo no appreciable changes after laser radiation. This process provides a possible basis for its application in micropatterning of transparent materials using excimer lasers.

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Optical materials such as fused silica, calcium fluoride and sapphire exhibit no appreciable absorption in the near-UV region and hence are transparent to the common excimer laser wavelengths (192, 222, 248, 308 and 354 nm). This makes it difficult to directly process these transparent materials using such a laser system. Earlier work has reported that surface modification can be produced on fused silica under either long-wavelength 10.6-μm radiation [1] due to resonant response or 0.53-μm radiation of ultrashort 2-ns pulses [2] due to the formation of laser-driven shock waves. However, other workers [3] have shown that damage patterns such as cracks and bubble-like damage can be produced on fused silica, crystalline quartz and borosilicate glass under laser irradiation at 1064 and 532 nm, wavelengths at which all these materials are transparent. The damage induced in these materials is explained in terms of mechanisms involving either electron avalanche, multiphoton absorption, inclusion heating or bond breaking. It is interesting to note that the surface layer of silicon dioxide can be modified by ion bombardment, resulting in a rather strong absorption in the near-UV due to the presence of a high concentration of Si-Si bonds in this layer [4]. This modification of the $SiO_2$ surface also provides an approach to subsequent direct processing of the $SiO_2$ surface by excimer lasers. In this study, an alternative method is proposed to form highly resolved micropatterns in transparent oxide layers using a conventional excimer laser. The $SiO_2$ layer with a thickness up to 440 nm is first deposited onto a single-crystalline silicon wafer. A KrF excimer laser beam is then directed perpendicular to the $SiO_2$ surface and micropatterns can be formed on the oxide surface. The pattern size can be controlled by adjusting either the laser parameters or the oxide layer thickness. The mechanisms involved in the formation of these micropatterns are discussed in terms of laser-induced melting of the $SiO_2/Si$ interfaces and accompanied phenomena.

## 1 Experiment

Figure 1 shows the experimental setup employed in this study. A Lambda Physik KrF excimer laser (LPX50) operating at a wavelength of 248 nm and producing a pulse of approximately 23-ns FWHM (full width half maximum) duration was used. The maximum pulse repetition rate was 30 Hz and the maximum pulse energy available was up to 300 mJ. The laser beam first passed through an aperture and was then directed by a reflecting mirror. To reach the required fluence, a quartz plano-convex lens with a focal length of 200 mm and a diameter of 500 mm, was used to focus the beam onto the sample surface. The samples were $SiO_2$ films of different thicknesses varying from 24 to 440 nm which were grown on P-doped *n*-type (100) silicon wafers with a resistance of 8–12 Ω cm using either conventional thermal oxidation or rf sputtering. The samples to be irradiated were placed in air with the oxide surface perpendicular to the incident laser beam. The micropatterns produced on the samples after laser irradiation were characterized by a NanoScope Dimension 3000 atomic force microscope (AFM, Digital Instruments,

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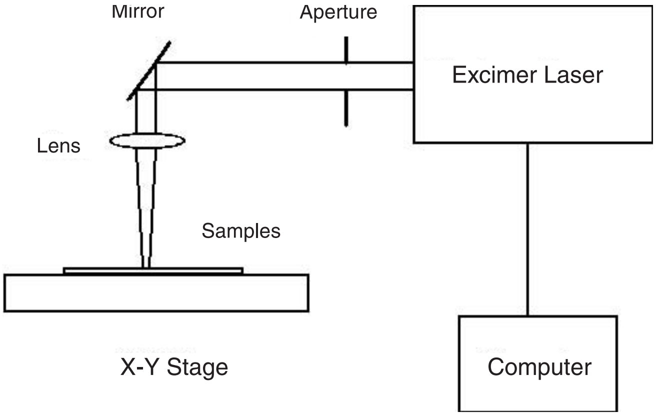


**Fig. 1.** Experimental setup employed in this study

Santa Barbara, CA), a Hitachi S-4100 field emission scanning electron microscope (SEM), and a Digilab FTS-6000 Fourier transform infrared (FTIR) spectrometer.

## 2 Results and discussion

Figure 2 shows the AFM images of micropatterns formed on a rf-sputtered 85-nm thick oxide layer when $SiO_2/Si$ is irradiated by a KrF (248 nm) laser beam with a single pulse at a fluence of $724\,mJ/cm^2$. Figure 2a–c shows the two-dimensional, three-dimensional and cross-sectional profiles, respectively. It was found that a well-defined ripple pattern could be produced on the $SiO_2$ layer surface after the normal incidence of irradiation with this KrF laser beam. The line scan in Fig. 2c shows the ripple period and vertical variation to be approximately 3.0 μm and 90 nm, respectively.

The size of the pattern produced on the transparent $SiO_2$ layer is expected to vary with laser fluence and pulse number. Figure 3 shows the mean ripple period as a function of laser fluence for a rf-sputtered 120-nm $SiO_2$ layer on Si irradiated by a single KrF laser pulse. Within the range used, the mean ripple period increases almost linearly from 1–6 μm with increasing laser fluence. Figure 4 indicates the ripple period as a function of pulse number for a rf-sputtered 85-nm $SiO_2$ layer on Si irradiated by a KrF laser beam at $724\,mJ/cm^2$. It can be observed that the ripple period grows rapidly with the first few pulses and then tends towards a steady-state value after about 20 pulses. This level will be different for different levels of fluence, and suggests that at certain fluence, there always exists a critical pulse number after which the ripple period saturates. Similar phenomena have emerged on laser-irradiated polymers [5] and semiconductors [6].

Figure 5 shows the ripple period as a function of oxide thickness for the $SiO_2$ layers thermally grown on Si irradiated by KrF laser pulses at a fluence level of $724\,mJ/cm^2$. As the oxide thickness increases from 24 to 440 nm, the ripple period increases linearly from 1.6 to 15 μm. The relationship of the ripple period and the oxide thickness can be formulated as

$$\Lambda = \Lambda_0 + K h_f \,, \qquad (1)$$

where $\Lambda$, $\Lambda_0$, $K$, and $h_f$ are the ripple period, initial ripple period on the bare Si surface, coefficient, and oxide thickness, respectively. $\Lambda_0$ and $K$ are found to be 1.0 and 26.7 μm, respectively. During irradiation, since the molten Si is covered by oxide layers with different thicknesses, the ripple structures generated at the interface may be affected by different strengths of the mechanical forces due to these different thicknesses. This provides a novel approach to controlling pattern size of the microstructures, which could have pronounced advantages over adjustment of laser parameters, since the latter are often difficult to control precisely.

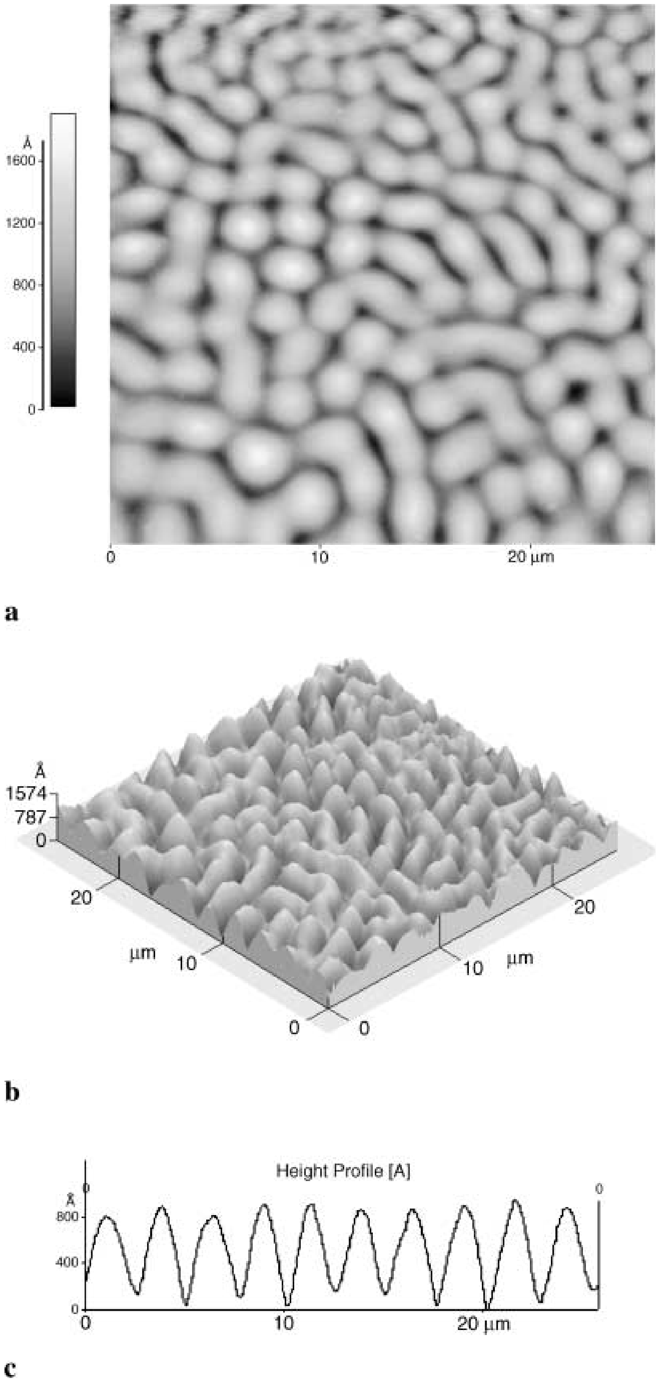


**Fig. 2.** AFM images of micropatterns formed on a rf-sputtered 85-nm thick oxide layer when $SiO_2/Si$ is irradiated by a KrF laser beam with a single pulse at a fluence of $724\,mJ/cm^2$: **a** two-dimensional, **b** three-dimensional, and **c** cross-sectional traces

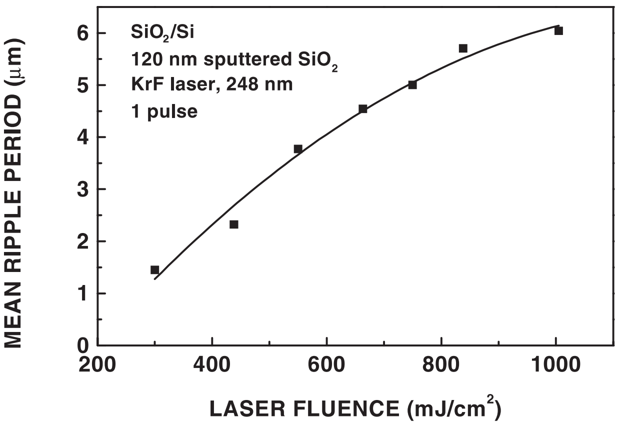


**Fig. 3.** Ripple period as a function of laser fluence for a rf-sputtered 120-nm $SiO_2$ layer on Si irradiated by a single KrF laser pulse

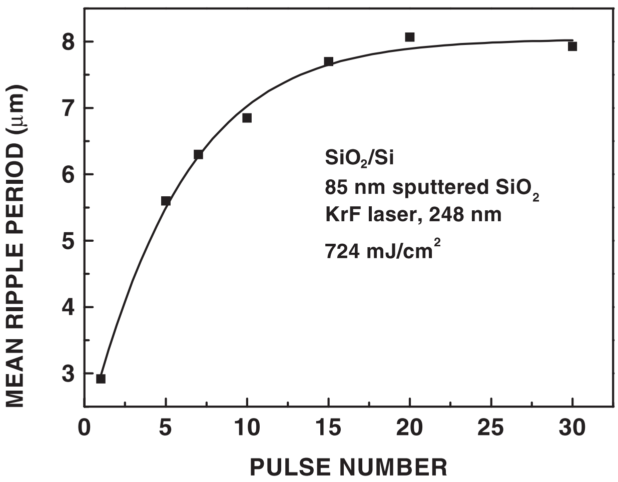


**Fig. 4.** Ripple period as a function of pulse number for a rf-sputtered 85-nm $SiO_2$ layer on Si irradiated by a KrF laser beam at a fluence of 724 mJ/cm$^2$

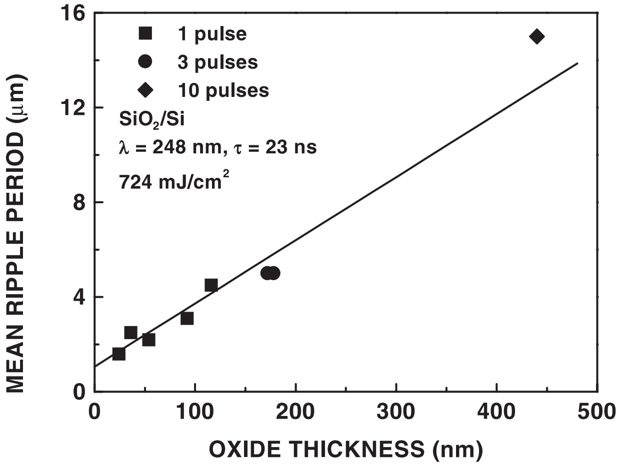
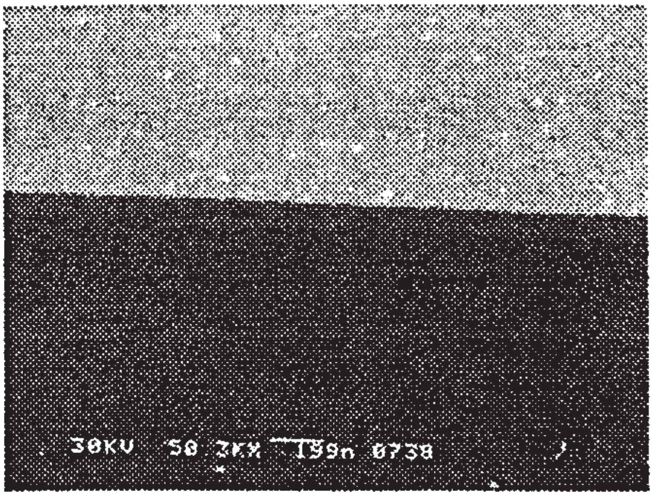
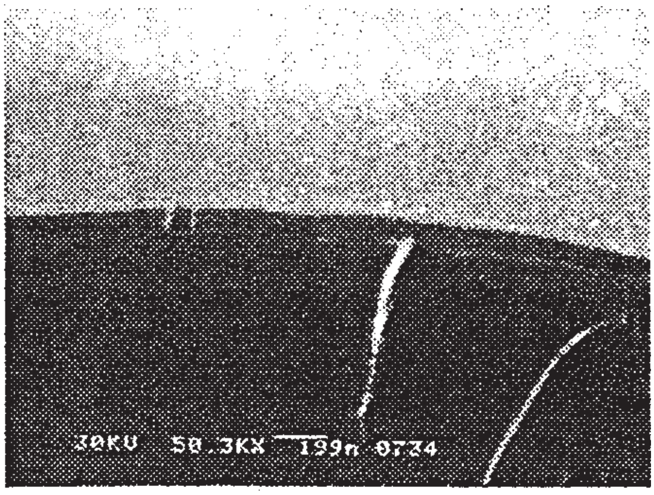


**Fig. 5.** Ripple period as a function of oxide thickness for the $SiO_2$ layer thermally grown on Si irradiated by a KrF laser beam at a fluence of 724 mJ/cm$^2$



**a** Before laser irradiation



**b** After laser irradiation

**Fig. 6a,b.** Cross-sectional SEM micrographs of a 116-nm thermally grown $SiO_2$ layer on Si before (**a**) and after (**b**) a single KrF laser pulse of irradiation at 724 mJ/cm$^2$

The initial seeding of the ripple pattern and the change of oxide surface profile can be identified in Fig. 6 which shows cross-sectional SEM micrographs of a 116-nm $SiO_2$ layer thermally grown on Si before and after a single KrF laser pulse of radiation at a fluence of 724 mJ/cm$^2$. From this figure, it can be observed that although the laser irradiation causes the melting of the interface, it is not sufficiently strong to cause a complete melting of the $SiO_2$ adjacent to the interface. Therefore, the $SiO_2/Si$ interface is still as sharp as before radiation although the interface protrudes towards the $SiO_2$ layer showing the interface profile has been changed. Close scrutiny of the samples reveals no trace of film cracking after laser irradiation, suggesting that the change in the surface profile of the thin $SiO_2$ layer may be a result of elastic strain following laser-seeded oscillation of the molten substrate surface.

If the laser fluence is sufficiently large, heat conducted from the interface to the oxide layer can cause melting or ablation of the oxide adjacent to the interface. Laser-induced temperature rises for double-layered structures can be simulated using the one-dimensional heat flow equation, which is described at length elsewhere [7]. Figure 7 shows the surface temperature as a function of time for a 40-nm $SiO_2$ layer on Si irradiated by a 23-ns KrF laser pulse at different fluences. The substrate temperature is taken to be 300 K. It is observed that when the fluence is increased to 0.65 J/cm$^2$, onset of melting occurs in the oxide surface. The energy with a fluence of 1 J/cm$^2$ is, however, large enough to cause complete melting of the oxide layer and produce latent heat to maintain a molten state for a short period of time (about 11 ns) after the laser pulse ends. With laser fluences further increased to 2 J/cm$^2$, the oxide surface begins to evaporate but the laser energy is not enough to produce the latent heat for vaporization, thereby leaving a flat top in the temperature profile. Upon cooling, laser irradiation at 2 J/cm$^2$ causes the oxide layer to remain molten much longer than that at 1 J/cm$^2$ as the increased latent heat accumulated from the former will then be released. Therefore, if laser fluence is properly selected, melting of the oxide adjacent to Si can be caused by the heat conducted from the molten interface. The molten oxide layer will thus assume the same profiles of the micropatterns induced on the molten substrate surface upon cooling, which has been well demonstrated in our experiment [7]. These physical processes provide a potential approach to micropatterning of transparent materials using excimer lasers. A highly absorbing layer which acts as a mask can be deposited on one side of the transparent material. When the excimer laser beam with a sufficiently large fluence passes through another side of the transparent material to arrive at its interface with the highly absorbing layer, laser-induced melting occurs at the interface. If the interface temperature rises to well above the melting or ablation point of the transparent material, a thin layer of the transparent material adjacent to the highly absorbing layer will melt or be ablated and eventually take on the same pattern as the mask layer.

It is essential to monitor the effect of laser irradiation on the properties of the processed material. Figure 8 presents the IR transmission spectra of a rf-sputtered 30-nm $SiO_2$ layer on Si before and after a single KrF laser pulse of radiation at a fluence of 1 J/cm$^2$. Both the spectra before and after laser irradiation contain the most prominent Si-O stretching mode peaks. The bending and rocking modes are less distinct. Both the peak positions of the stretching mode before and after laser irradiation are located at 1077 cm$^{-1}$, which are characteristic for $SiO_2$ films. This suggests that structural properties of the $SiO_2$ layer can remain unchanged after such a laser irradiation operation.

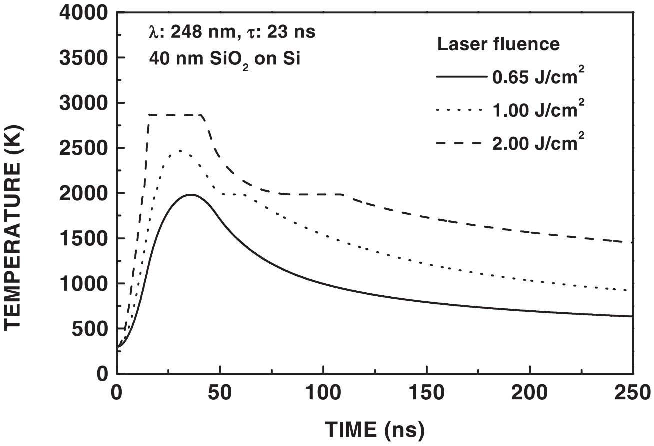


**Fig. 7.** Surface temperature as a function of time for a 40-nm $SiO_2$ layer on Si irradiated by a 23-ns KrF laser pulse at different fluences

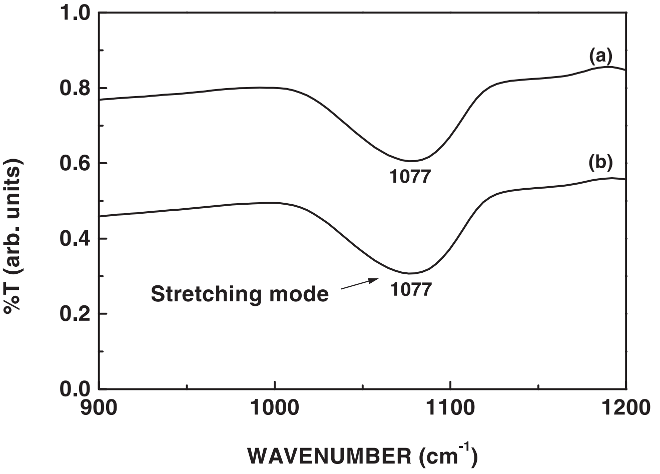


**Fig. 8a,b.** IR transmission spectra of a rf-sputtered 30-nm $SiO_2$ layer on Si before (**a**) and after (**b**) a single KrF laser pulse of radiation at a fluence of 1 J/cm$^2$

## 3 Conclusion

Highly resolved micropatterns induced in transparent materials were investigated using a KrF excimer laser and discussed in terms of laser-induced interface superheating. It was found that a well-defined ripple pattern with micrometer-order periodicity and nanometer-order roughness could be generated on the $SiO_2$ layer surface for a $SiO_2$/Si layered structure to be irradiated by a KrF laser beam. The ripple periodicity was observed to increase almost linearly with laser fluence, tending towards a saturation value after 20 pulses of radiation. The ripple periodicity was also found to have a linear dependence on the oxide layer thickness, which provides an alternative approach with pronounced advantages over adjustment of laser parameters for controlling the pattern size of the microstructures. SEM analysis demonstrated that the original seeding of ripples occurred at the molten Si surface and the elastic $SiO_2$ layer exactly followed the interface rippling to change its profile. Simulation of laser interaction with double-layered structures indicated that the oxide layer could melt or be ablated due to interface superheating when it was deposited on a highly absorbing Si substrate. IR analysis exhibited no appreciable changes caused in structural properties of the oxide layer after laser radiation at a fluence of 1 J/cm$^2$. This process can be extended to micropatterning of the transparent material using excimer lasers through its superheated interface with a highly absorbing mask layer.

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