
RIPPLE FORMATION ON InP SURFACE IRRADIATED WITH FEMTOSECOND LASER

H.X. QIAN and W. ZHOU *
School of Mechanical and Aerospace Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798
* WZhou@Cantab.Net

H.Y. ZHENG
Singapore Institute of Manufacturing Technology, 71 Nanyang Drive, Singapore 638075

Single crystalline InP was ablated with linearly p-polarized femtosecond laser in air. Ripples with orientation parallel to the laser polarization direction were formed at low laser fluence. Analyses by EDX reveal In oxides and P oxides on the surface. Micro Raman spectroscopy of the laser irradiated surface indicates presence of stresses and possible formation of InP nanocrystals due to the femtosecond laser irradiation.

Keywords: Femtosecond laser; InP; ripple; oxidation; Raman spectroscopy

1 Introduction

Laser-induced periodic surface structure may have important engineering applications, e.g., as filters or gratings. Since such surface structure was first observed by Birnbaum in 1960’s, laser-induced surface ripples have been reported by many other researchers in a wide range of materials. The ripple formation is commonly believed to be caused by interference of the incident laser with some form of surface scattered wave. In many cases, the ripple period is close to laser wavelength, but subwavelength ripples have also been reported in recent years. Borowiec et al. reported ripples perpendicular to laser polarization direction on InP surface. In our study of InP surface irradiated with femtosecond laser, however, we observed ripples parallel to the laser polarization direction. In the paper, we discuss the seemingly contradictory results and report the results of further characterization of the laser irradiated surface by EDX and Raman spectroscopy.

2 Experimental

Undoped InP (100) wafer was irradiated in air using linearly p-polarised femtosecond laser with wavelength of 775 nm, pulse duration of 150 fs and repetition rate of 1 kHz. The wafer was placed at a position of 4 mm away from the focal plane of the focusing lens (focal length f = 50 mm). The corresponding nominal beam diameter at the off-focal

779
plane was 293 µm. The laser power was fixed at 51 mW to generate single pulse energy of 51 µJ and laser fluence of 76 mJ/cm². The laser beam was always directed perpendicularly onto the sample surface but the number of pulses was varied.

After laser ablation, the surface morphology was examined using scanning electron microscope (CamScan S360) and atomic force microscope (Shimadzu SPM-9800). The chemical composition was studied using energy dispersive X-ray spectroscopy (EDX). Micro Raman spectroscopy was carried out at room temperature using a Renishaw system with a HeNe laser to study stress and structural change within the irradiated areas. A backscattering geometry was adopted for all the Raman measurements.

3 Results and Discussion

Periodic surface structures were found to appear after the femtosecond laser irradiation, as shown in Figure 1. With the number of laser pulses increasing from 50 (Figure 1(a)) to 300 (Figure 1(b)), the ripples are always aligned parallel to the laser polarization direction. 3D morphology of the ripples was obtained using AFM. It can be seen from the AFM image in Figure 2 that depth of the ripples is about 1000 nm for the smallest number of pulses used (50). When the number of pulses increased from 50 to 300 and above, the ripples became even rougher and thus increasingly difficult for the AFM characterization.

It is interesting to note that the ripples are parallel to the laser polarization direction, because this is in strong contrast to the observation by Borowiec et al. 12 and Bonse et al. 10,13 that ripples on the same type of material are perpendicular to laser polarization direction. However, our results are not necessarily contradictory to those by others 10,12,13. In fact, they could be complementary to the previous results and thus help to gain a more complete picture of the complex ripple formation process.

Although Bonse et al. 10,13 reported formation of ripples perpendicular to laser polarization, they also observed the morphological change from the ripples to so-called “grooves” when the number of laser pulses increased. It should be noted that the grooves were in fact parallel to laser polarization (see Fig. 4(d) in Ref. [10] and Fig. 6 in Ref. [13]). In the present study, when the number of pulses increased to 3000, another type of periodic structure appeared on surface and their orientation is perpendicular to the laser polarization direction, as shown in Figure 3.
Figure 1 SEM images of ripples on InP (100) irradiated with number of laser pulse of (a) 50 and (b) 300 at laser fluence of 76 mJ/cm$^2$. The arrows indicate the laser polarization direction.

Figure 2 AFM image showing ripple morphology on InP (100) irradiated with 50 laser pulses at laser fluence of 76 mJ/cm$^2$.

Figure 3 SEM image of InP (100) irradiated with 3000 laser pulse at fluence of 76 mJ/cm$^2$ showing appearance of another type of periodic structure perpendicular to the laser polarization direction (arrowed) in addition to the parallel ripples.
When the number of laser pulses is small at 50, the ripples have period of roughly three quarters of the laser wavelength (Figure 1(a)). However, the period tends to increase with the number of pulses and grows slightly above the laser wavelength at 3000 pulses (Figure 3). Period for the structures perpendicular to the polarization direction is much broader. The mechanisms of the ripple formation are complex and warrant further study.

Raman scattering spectra are presented in Figure 4. The raw surface exhibits a strong longitudinal optical (LO) phonon at 345 cm\(^{-1}\), and the second-order Raman peak 2LO at 687 cm\(^{-1}\). In the chosen excitation geometry, the transverse optical (TO) peak is forbidden by the Raman selection rules in InP (100)\(^{14}\). Therefore, TO is indistinguishable on the raw surface. After laser irradiation, both the TO mode and LO mode were observed. The TO was found to be near 302 cm\(^{-1}\), and the LO around 344 cm\(^{-1}\).

The linewidth of the LO phonon broadens and the broadening becomes asymmetrical towards the low frequency side. The broad features are induced by the coherence length of the phonons in the disordered material\(^{14}\). The presence of the LO and TO modes indicates the statistically distributed crystal orientations on the irradiated areas. The broadening of LO peak together with the appearance of TO mode might indicate the formation of InP nanocrystals\(^{15,16}\). The shift in the peak position reveals that femtosecond laser ablation induced stress on irradiated surfaces\(^{17}\).

Figure 4 Raman spectra for surfaces irradiated with different numbers of laser pulses at laser fluence of 76 mJ/cm\(^2\).
O concentration on the irradiated surfaces was analyzed using EDX. The O concentration is almost 0 for the raw surface but increases substantially after laser irradiation, as shown in Figure 5. It is very interesting to observe two distinct regimes from the figure. In the regime of small number of pulses (50-3000) for ripple formation, O concentration fluctuates around 10-20 at.%. Above 3000 pulses, O concentration rises drastically and arrives towards a plateau at approximately 70 at.%. The abrupt increase in O is accompanied by surface morphological evolution from ripples to flower-like structures. Previous study\textsuperscript{18} reported the formation of flower-like structures on InP and showed by XPS that they are composed of In oxides and P oxides redeposited onto the surface. The EDX analyses lend further support to the XPS study and indicate the formation of oxides even after a small number of laser pulses.

4 Conclusion

Ripples with period equal to three quarters of laser wavelength formed at low laser pulses of 50. The period increases with laser pulses and grows slightly above the laser wavelength at 3000 pulses. The ripples are parallel to the laser polarization direction, but much broader groove-like periodic structures perpendicular to laser polarization direction also formed at large number of pulses. Raman spectroscopy indicates presence of stresses and possible formation of InP nanocrystals due to the femtosecond laser irradiation.

The EDX analyses reveal In oxides and P oxides on the surface.

References