Study of phase change LIPSS formation in Si by fs-resolved microscopy

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Abstract: We report the formation of amorphous-crystalline gratings in c-Si by fs laser irradiation at different wavelengths, repetition rates and angles of incidence. Using fs-resolved microscopy, we reveal the phase transformation dynamics of these structures.

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1. Introduction

One of the most interesting properties of Silicon for the electronics industry is that it can be cycled between the crystalline and amorphous phase. The phase change can be induced by laser irradiation, with the amorphous phase being obtained by a melting process followed by fast quenching, while recrystallization is induced by laser thermal annealing. The use of ultrashort pulses (pico- and femtosecond) allows triggering non-thermal phase changes which may lead faster amorphization and recrystallization processes [1-2].

Another process that can be triggered in Si is the formation of Laser-Induced Periodic Surface Structures (LIPSS). This self-organization phenomenon is produced by an interference process between the incident laser light and a scattered surface wave, yielding topographic grating structures with an orientation typically perpendicular to the laser polarization and periods in the order of the laser wavelength [3]. While LIPSS have been observed mainly in the ablation regime [4], recently *Katsumata et al.* have observed self-organized periodic amorphous-crystalline structures [3] in thin GeTe films after irradiation with multiple femtosecond laser pulses [5].

2. Result

In this work, we combine both phenomena (laser-induced phase changes and LIPSS) to fabricate amorphous and crystalline grating structures. Our strategy for static irradiation is to use a first femtosecond laser pulse to form a homogeneous, ultrathin amorphous surface layer, whereas a second pulse triggers amorphous-crystalline LIPSS formation. The grating formed has a lateral extension of a few micrometers and a period close to the laser wavelength, which can be resolved by optical microscopy due to the different optical properties of the two structural phases of Si.

Figure 1. Optical micrographs of lines written under different laser (370 fs, 500 kHz and 1030 nm (a), 100 fs, 100 Hz, 800 nm (b) and 100 fs, 100 Hz, 400 nm (c)) and irradiation conditions (normal incidence (a), incident at an angle of 53° (b,c)). The beam was horizontally polarized. (d) 2D amorphous-crystalline grating written under the same conditions as in (a).

We have extended this static experiment into extended lines of gratings (Fig. 1a-c, Left) by scanning the laser beam over the surface. In order to identify the adequate spatial pulse overlap in scanning conditions we have written lines at different scan velocities. We found that a well-defined spatial pulse overlap needs to be kept in order to ensure the formation of pure amorphous-crystalline structures and that any deviation from it leads either to inhomogeneous or ablative structures. We also demonstrate that this irradiation strategy can be extended to other laser repetition rates and wavelengths (Fig. 1b-c, 100 Hz, 100 fs, 800 (b) and 400 nm(c)), and that the grating period can be tuned by changing the angle of laser beam incidence (Fig. 1b-c, 53°). Choosing a certain spacing between written lines allows extending these lines into large-area 2D structures by interconnecting ripples of each line (Fig. 1d, Right)

We will also present first results on the temporal and spatial dynamics of the ripple formation mechanism, obtained by means of a fs-resolved microscopy system. The time-resolved images (Fig. 2a) correspond to snapshots of the transient reflectivity of the sample surface in scanning conditions recorded at a certain delay time after the arrival of the pump pulse. The reflectivity profiles of these images (Fig. 2c-b) reveal that ultrafast melting and rapid solidification are involved in this type of LIPSS formation. In particular, our results are able to resolve the birth of a new ripple (Fig. 2c, “infinite” arrow), beginning with an ultrafast reflectivity increase, consistent with melting (Fig. 2b, 300 fs), and ending with rapid solidification after a few ns (Fig. 2c, 5 ns). These findings shed new light on the ongoing vivid debate about the formation mechanism of LIPSS in Si, and the technique employed here might contribute to clarify the formation mechanism of other LIPSS types.

Figure 2. (a) Snapshot of the surface reflectivity 300 fs after the arrival of the pump pulse (800 nm, 100 fs, horizontally polarized, angle of incidence 53°) in writing conditions of a line of amorphous-crystalline gratings (b) Reflectivity profiles (b-c) of the irradiated region at different delays The arrow indicates the position of the new single ripple formed (“infinite”).

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