Employing Reactive Gas-phase Etch Precursors to Mitigate Redeposition During Ultrashort Pulsed Laser Ablation

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Ultrashort pulsed laser ablation (UPLA) enables material removal rates three to six orders of magnitude greater than those of LMIS focused ion beam techniques. However, while UPLA purports to be precise, deterministic, and a-thermal, mill quality is often compromised by surface debris and the redeposition of material generated during ablation. It is well known that samples processed in vacuum suffer less surface debris than those ablated in inert gaseous atmospheres due to an increase in the mean free path of particles ejected during ablation [1]. However, even in vacuum, ablated material tends to redeposit inside milled features (e.g. at the trailing edge of the scan pattern), thereby partially refilling and deforming features as they are machined. While other researchers have investigated UPLA in inert "assist" gases [2], we present the first evidence that samples ablated in reactive gaseous atmospheres can show a dramatic improvement in cut quality due to a reduction in redeposition. Material removal rate dependencies on process parameters indicate that that the material ejected during UPLA reacts with precursor molecules to form volatile compounds, and that the relevant reaction occurs primarily in the gas phase, in contrast to gas-assisted electron and ion beam induced etching where the process is mediated by precursor molecules adsorbed to the sample surface.

UPLA was performed at room temperature using a Ti:Sapphire chirped pulse amplification laser with a center wavelength of 775 nm, a pulse duration of 150 to 200 fs, and a repetition rate of 1 kHz. The precursor and sample were XeF_2 and z-cut SiO_2. Control experiments were performed in vacuum and in a N_2 atmosphere. Fig. 1 shows a set of nominally square pits machined in SiO_2 in high vacuum and at XeF_2 pressures of 0.3, 0.5, 1, 2 and 4 torr. The stage was scanned in a serpentine fashion, starting in the top left-hand corner of each box. The sample region milled in high vacuum contains a large amount of material redeposited along the upper edge and sidewalls of the ablated feature. The amount of recast material is seen to decrease with increasing XeF_2 pressure. This improvement in feature geometry was not observed in equivalent experiments performed in a N_2 atmosphere and is ascribed to volatilization of species ejected during UPLA. The formation of volatiles can proceed through a number of mechanisms including (i) the dissociation of gas-phase and adsorbed XeF_2 to generate reactive species that can volatilize material ejected during UPLA, and (ii) the generation of Si-containing species (other than SiO_2) during ablation that can react with gas-phase and adsorbed XeF_2. The material removal rates indicate that adsorbates play a negligible role in the volatilization process which is mediated primarily by gas-phase precursor molecules. The volatilization mechanisms will be discussed in detail in the light of systematic experiments performed as a function of process parameters.

[1] D. Bleiner, T. Lippert, J. Appl. Phys. **106,** 123301 (2009)
[2] G. M. Robinson, M. J. Jackson, J. Mater. Eng. Perf. **15**, 155 (2006)

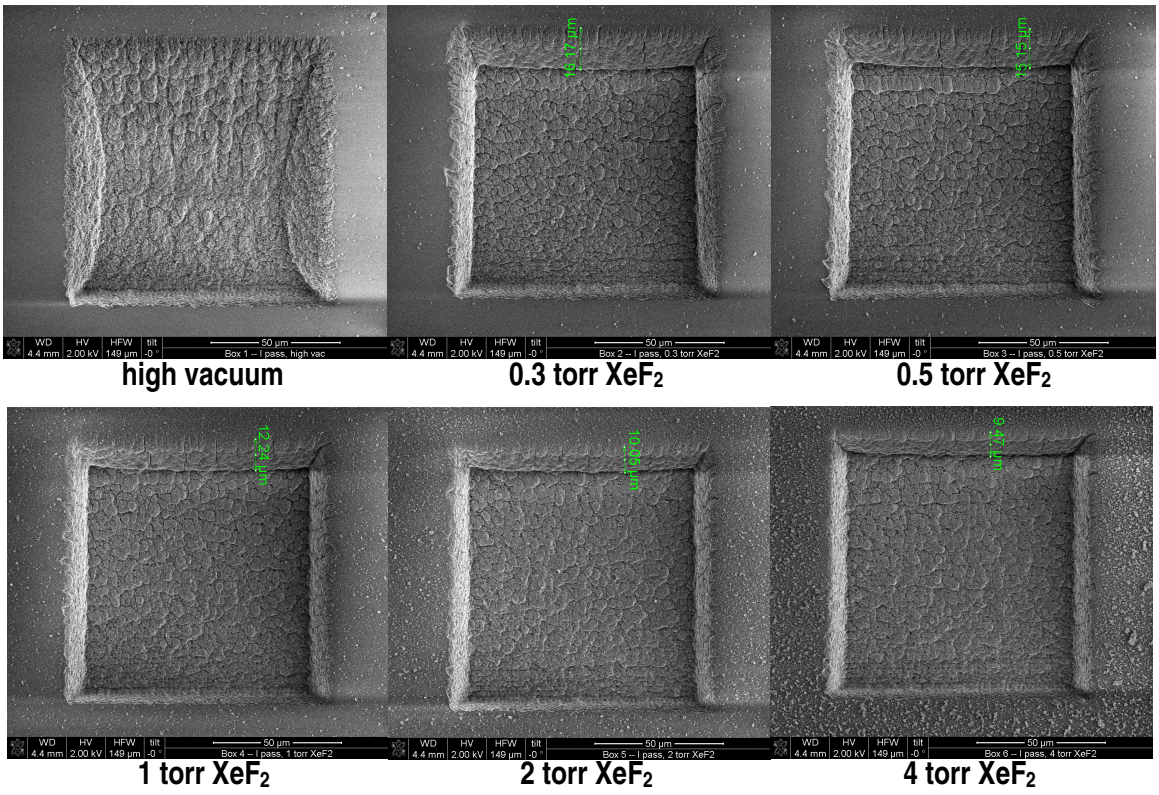

Fig. 1 Top-down scanning electron micrographs of 100 μm^2 boxes machined in SiO_2 at a number of XeF_2 pressures.