Hierarchical microstructures with high spatial frequency laser induced periodic surface structures (HSFLs) possessing different orientations created by high-fluence femtosecond laser ablation of silicon in liquids

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Abstract

Phenomena widely known as laser induced periodic surface structures (LIPSS) on silicon typically include three features; (1) high spatial frequency laser induced periodic surface structures (HSFLs) can be achievable at low laser fluence in the range of 0.1~1 J/cm^2 near ablation threshold, (2) the period of HSFLs is greatly influenced by the pulse number used for ablation and (3) the direction of HSFLs generated by laser ablation both in air and liquids is perpendicular to the light polarization. This work provides some new findings for LIPSS formation by femtosecond laser (pulse duration of 457 fs, wavelength of 1045 nm, and repetition rate of 100 kHz): (1) Most importantly, silicon HSFLs (Si-HSFLs) with periods of 110-200 nm can be generated for the first time by laser ablation in liquids (LALs) at the laser fluence (85 J/cm^2) much higher than ablation threshold, which enables creating hierarchical microstructures. (2) High repetition rate pulse irradiation is another important factor for the HSFL formation. (3) The period of Si-HSFLs is independent of the scanning speed (0.1, 0.5, 1 and 2 mm/s), line intervals (5, 15 and 20 μm) of scanning lines and scanning directions (perpendicular or parallel to light polarization). (4) When microstructures with height gradients are produced, the directions of some HSFLs are no longer perpendicular to that of light polarization to tilt clockwise or anticlockwise randomly with maximal deviation angle of 50°. Raman spectra and SEM characterization jointly confirm the underlying correlation of surface melting and nanocapillary waves with the formation of Si-HSFLs. No production of HSFLs by laser ablation in air indicates that moderate melting facilitated with ultrafast liquid cooling is beneficial for the formation of HSFLs by LALs. Based on our findings and deduced from previous reports, the formation mechanism of HSFLs at high fluence is proposed, which is a combination of thermal melting with the cooperation of ultrafast cooling in liquids and modulation of the molten layers into ripples and nanotips by surface plasmon polaritons (SPP) and second-harmonic generation (SHG). Furthermore, we attribute the modulation of Si-HSFLs direction to the nanocapillary waves and the localized electric field generated by the large particles.