Surface nanostructuring by short-range propagating surface plasmon excited with few-cycle femtosecond laser pulses

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Laser-induced periodic surface structure (LIPSS) with a period *d* of 1/10-1/4 of laser wavelength on solid surfaces are formed with superimposed multiple shots of femtosecond (fs) laser pulses below a single-shot ablation threshold. Recently, few-cycle laser pulses with a pulse duration of 7 fs were observed to be able to form much finer LIPSS on a diamond-like carbon (DLC) film than that formed by the 100-fs pulses [1]. Moreover, it was shown that the physical processes are the generation of a high-density electron layer with a thin thickness and the excitation of short-range surface plasmon polaritons (SR-SPPs) [2,3]. However, because the formation of finer LIPSS was observed only on the DLC film, the detailed mechanism has never been understood yet. In this paper, we report the formation process of the finer LIPSSs on metal and semiconductor surfaces with the 7-fs pulses.

For the ablation experiment, we used polished stainless steel (SS304) and *n*-type gallium arsenide substrates as targets. The linearly polarized 7-fs laser pulses output from a laser oscillator (650-1000 nm, 81 MHz, 420 mW) were focused on the targets with a reflective objective. The surface morphology of the irradiated target surfaces was observed with a scanning electron microscope. The bonding structural changes of the irradiated targets were measured with micro-Raman spectroscopy.

The nanostructures with d = 60-110 nm were formed on the SS304 surfaces, as the surfaces were modified to Fe₂O₃. In increasing the peak power density *I*, *d* and the Raman signals indicating Fe₂O₃ increased. The period $D = \lambda_{spp}/2$ was calculated as a function of the density N_e of the electrons in the thin layer on the Fe₂O₃, where λ_{spp} is the surface plasmon wavelength. The results show, decreasing the thickness of the high-density electron layer, *D* decreases. The model calculation is in good agreement with the experimental results. These results show that SR-SPPs excited with few-cycle laser pulses induce the formation of finer periodic nanostructure on stainless steel surfaces.

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