§18. Mechanism of Nanostructure Formation on Metal Surface Induced by Intense Short Pulse Laser

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The periodic grating structures structures have been found in insulators, semiconductors, and metals. For metals irradiated with linearly polarized femtosecond laser, the self-organized grating structures on the metal surface are oriented perpendicular to the laser polarization. The interspaces of the grating structures are shorter than the laser wavelength and depend on laser fluence. However, the self-organization mechanism of the grating structures remains under investigation. The structures depend on material properties and laser parameters (wavelength, fluence, pulse duration, number of pulses, etc.). Even for the same metal, not all experimental results reported in the literature can be directly compared because of experimental differences, for example, in the laser parameters. For discussing the mechanism, a greater amount of systematic experimental data is now needed. One approach to gather systematic experimental data is to investigate the relation between the metal type and the interspaces of grating structures. Another approach is to investigate the relation between the ablation rate and the interspaces for typical metals.

We have reported that the grating structure interspaces depend on laser fluence for  $Cu^{1}$  irradiated with <100-fs laser pulses and for several metals<sup>2</sup>) irradiated with 160-fs laser pulses. In the laser fluence ranges where self-organized grating structures are formed, the interspaces of the grating structures are shorter than the laser wavelength of 800 nm. The interspaces increase up to 680 nm as laser fluence is increased. We have reported that the laser fluence dependence of the grating structure interspaces is the same for Ti, Pt, Mo, and W metals on which the self-organized structures are formed<sup>2),3).</sup> We found that, for these metals, the ratio of the grating structure interspaces to the wavelength of the laser light is 0.85 at  $F_M$  (the upper limit of the laser fluence range in which the self-organized grating structures are formed). We explained this dependence of the interspaces on laser fluence on the basis of a parametric decay (stimulated Raman scattering) model<sup>4</sup>). In this model, surface plasma waves (SP waves) are induced at the interface between free space (air) and either laser-produced plasma or metal plasma by parametric decay, resulting in the decay of the incident laser light into an SP wave and a scattered electromagnetic wave. The wavelength of the plasma wave induced on the surface depends on only the plasma frequency (electron density) of the surface plasma. The ratio of the wavelength of the SP wave to that of the laser light changes from 0.5 to 0.85 for plasma frequencies in the range of  $0 < \omega_{\rm p}/\sqrt{2} < \omega_L$ , where  $\omega_p$  and  $\omega_L$  are the frequencies of the plasma and laser light, respectively. When the plasma electron density is dependent on laser fluence, the wavelength of the SP wave in turn depends on laser fluence. This model satisfactorily explains the results reported for Cu<sup>4</sup>) and Ti, Pt, Mo, and W<sup>2</sup>). These results indicate that the parametric decay model is valid in regard to only the dependence of the interspaces on laser fluence. At present, however, the body of experimental data is insufficient for verifying this model. The dependence of the interspaces on the wavelength of the incident laser can also be explained in this model.

In the experiment, we investigated the states of metal surfaces (Ti and Mo) irradiated with femtosecond laser pulses at wavelengths of 800 and 400  $\rm nm^{5)}$ . We evaluated the dependence of periodic structure interspaces on laser fluence at the different wavelengths. We also examined the dependence of ablation rate on laser fluence at the different wavelengths. We found a relation between the ablation rate and laser fluence at which self-organized periodic structures were formed. In addition, we evaluated the threshold of plasma generation to discuss the extent of the parametric decay model's validity.

In order to visualize the surface plasma wave induced by femtosecond laser, two-dimensional particle in cell simulation by using the code  $FISCOF^{6)}$  has been demonstrated for initially pre-formed plasma on a target. The dependence of grating interspace on the laser wavelength was investigated<sup>7</sup>). It is found that the grating interspace is found to be almost half of the laser wavelength in spite of different wavelengths. The dependences of the grating interspace on the laser wavelength in simulations are similar to those in experiments.

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