Mathematical Model of Nonlinear Laser Lithography

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Laser induced periodic surface structures (LIPSS) had been observed just five years after the invention of laser [1]. Among the numerous LIPSS techniques none of them could maintain long-range order [2]. However, it has recently been demonstrated that long order periodic surface structures can be produced using nonlinear laser lithography (NLL) [3]. Here, we present a mathematical foundation for NLL.

NLL process is realised by the interplay of three physical events, i.e., positive feedback, nonlinearity and negative feedback. Each infinitesimal point on the target surface behaves like an individual scatterer of the incident electric field, in a dipole radiation pattern proportional to local height. This coupled dipole interference pattern created by a laser pulse determines the surface shape seen by the next laser pulse (positive feedback). According to the electric field distribution created by this interference, the target material ablates (nonlinearly) and reacts with the surrounding molecules in the surrounding environment. This process is limited by exhaustion of either molecular reactive species in the immediate vicinity of the ablation region (negative feedback). Both in our experiments (Fig. 1(a)) and simulations (Fig. 1(b-f)) we used titanium as target material and oxygen as ambient reactive species.

Fig. 1 (a) The schematic of the experimental setup. (b) Simulation results of scanning along the x-direction with vertically polarised laser beam. (c) Simulation results of scanning along the x-direction with horizontally polarised laser beam. (d) Experimental realisation of simulations shown in (b). (e) Experimental realisation of simulations shown in (c). (f) Simulation results of scanning along the x-direction with circularly polarised laser beam.

In the scattered field calculations, we ignored higher order effects with \(1/r^2\) and \(1/r^3\) terms. We modelled the scattered field with the following equations, which define the total dipole radiation on the surface element \((x,y)\):

\[
E_{x,scat}(x,y) = \gamma \int_S h(x',y') (\cos^2 \theta E_x(x',y') - \cos \theta \sin \theta E_y(x',y')) \frac{e^{ikr}}{r} \, dx' \, dy' \tag{1}
\]

\[
E_{y,scat}(x,y) = \gamma \int_S h(x',y') (-\cos \theta \sin \theta E_x(x',y') + \sin^2 \theta E_y(x',y')) \frac{e^{ikr}}{r} \, dx' \, dy' \tag{2}
\]

where \(E_x(x',y')\) and \(E_y(x',y')\) are the incident electric field components, \(h(x',y')\) is the surface height profile, \(\cos \theta = \frac{y'-y}{r}, \sin \theta = \frac{x'-x}{r}, r^2 = (x-x')^2 + (y-y')^2\), and \(\gamma\) is constant. In simulations, to obtain linear polarisation, we set the other field component to zero, i.e., for \(x\)-polarisation, \(E_y(x',y')\) is set to zero. Similarly for circular polarisation, we used \(E_x(x',y') = e^{i\phi} E_0 / \sqrt{2}\) and \(E_y(x',y') = E_0 / \sqrt{2}\). Simulations with vertical and horizontal polarisations can be seen in Fig. 1(b) and (c), respectively. We observe that the structures always emerge parallel to the laser polarisation. Fig. 1(d) and (e) illustrate the experiments corresponding to Fig. 1(b) and (c), respectively. Simulation with circularly polarised laser can be seen in Fig. 1(f), which produces a hexagonal pattern.

In conclusion, we present a mathematical model for NLL. The simulation results agree with experiments. We can experimentally and theoretically create long-range highly ordered periodic lines with femtosecond lasers. With circularly polarised lasers we can even produce hexagonal structures with long-range order and uniformity.

References