Evidence of plasmonic nature of self-arranged surface nanostructuring after single femtosecond laser pulse irradiation

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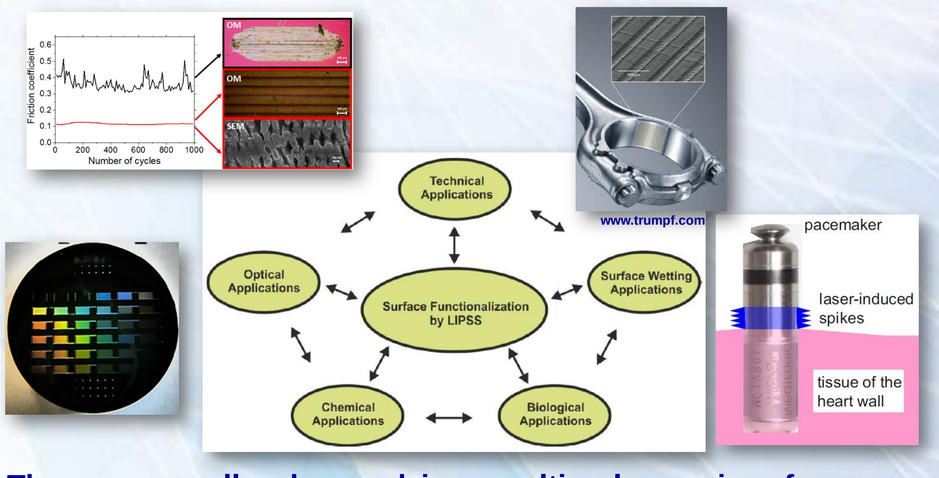
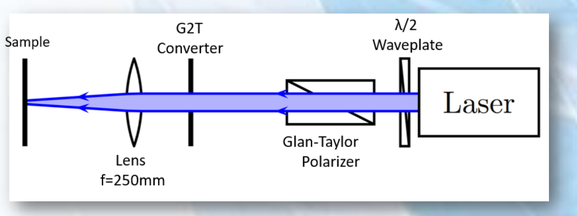
Motivation

Laser-induced periodic surface structures (LIPSS) have gained a broad spectrum of potential applications [1,2,3] .



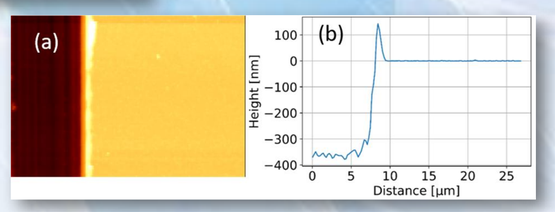
They are usually observed in a multi-pulse regime for many materials. However, what is the origin of their formation after a single pulse? How can we manipulate LIPSS morphology to fabricate desired structures?

Experimental setup

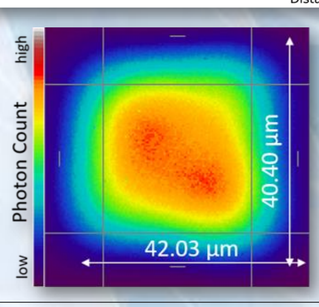


LASER PARAMETERS
- **Wavelength: 343 nm**
- **Pulse duration: 250 fs**

➢ **(a) top view and (b) height profile of a step edge on a gold sample.**



➢ **Flat-top laser beam profile on the sample.**



Theoretical approach

TTM+MD model [4,5]

$$\begin{cases} C_e(T_e)\dfrac{\partial T_e}{\partial t} = \nabla\left(K_e(T_e)\nabla T_e\right) - G(T_e - T_{lat}) + Q(\mathbf{r},t) \\ m_i \dfrac{d^2\mathbf{r_i}}{dt^2} = \mathbf{F_i} + \xi m_i \dfrac{d\mathbf{r_i}^{\mathrm{T}}}{dt},\ i = 1,\ldots,N_{atoms} \end{cases}$$

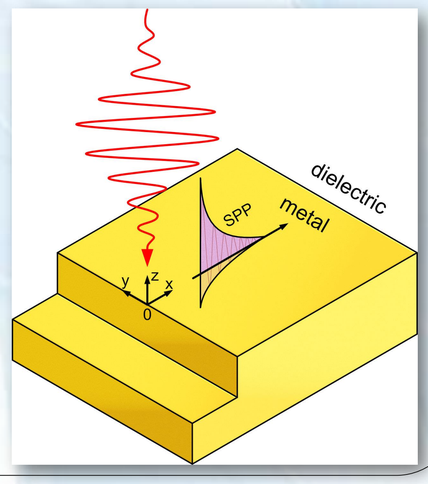
Laser source term

$$Q(\mathbf{r},t) = Q_{\text{las-las}}(\mathbf{r},t) + Q_{\text{las-SPP}}(\mathbf{r},t) + Q_{\text{SPP-SPP}}(\mathbf{r},t) \quad [6]$$

➢ **Schematic irradiation geometry with the excitation of surface plasmon polaritons (SPP) [7,8].**

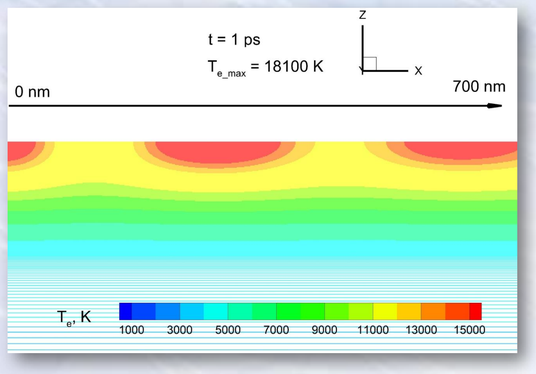
Interference:

laser fields + SPP fields

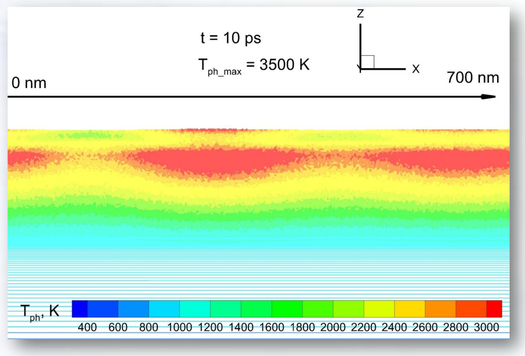


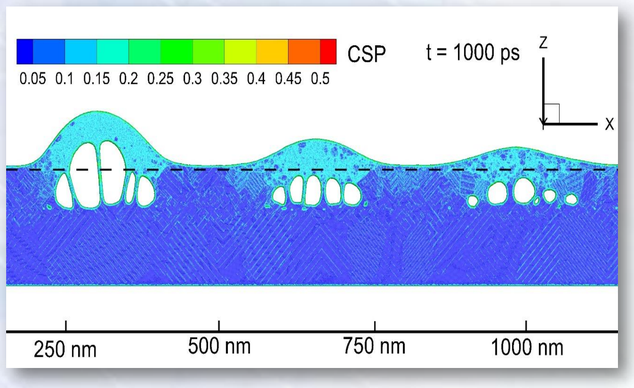
Theory & Experiment: comparison

Modulation of the electron temperature



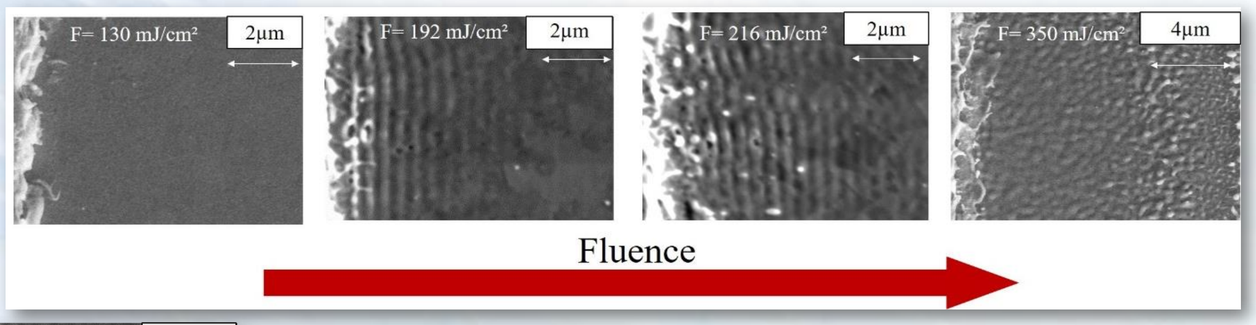
Modulation of the lattice temperature

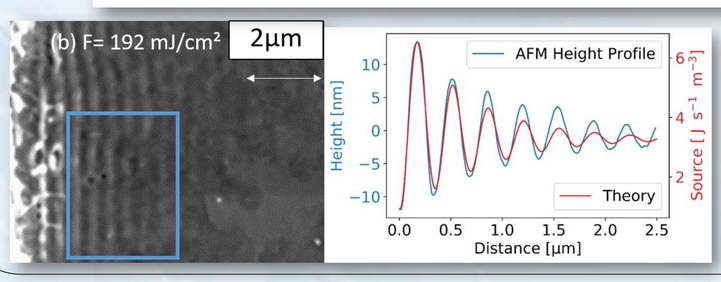




Morphology of periodic structures at 1 ns after the laser pulse

✓ **Structure height is determined by the interplay of SPP and laser fluence.**





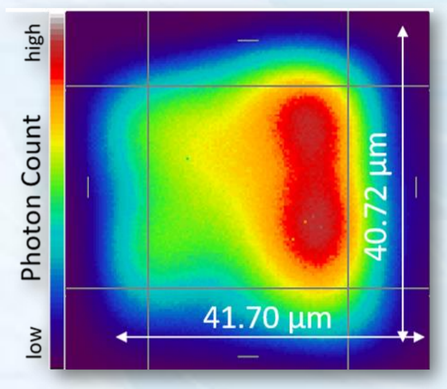
✓ **The measured periodicity matches theoretical predictions:**

$$\lambda_{\text{LIPSS}}^{\text{exp}} = (338 \pm 10)\,\text{nm}$$

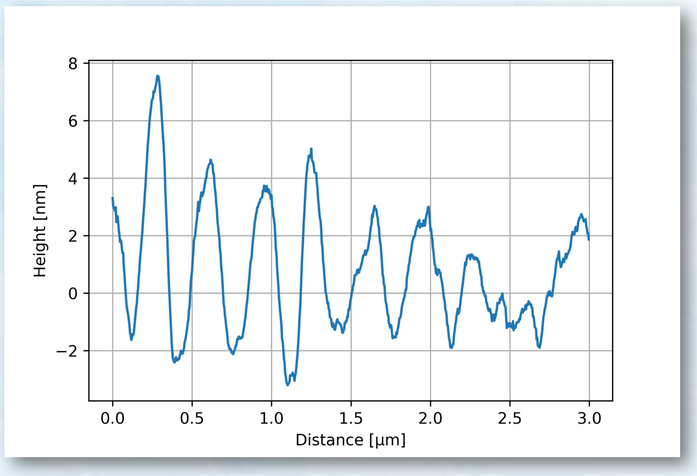
$$\lambda_{\text{SPP}}^{\text{theory}} = 344\,\text{nm}$$

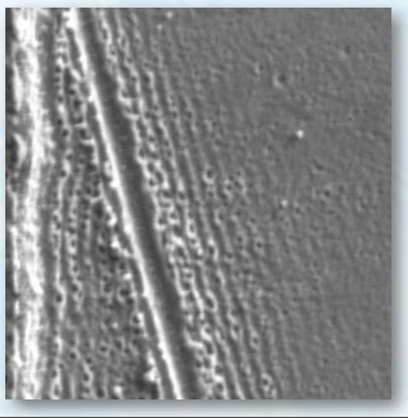
Design of LIPSS

➢ **Inhomogeneous laser beam profile on a sample.**



➢ **The height profile of LIPSS**





✓ **Influence of laser beam profile on LIPSS morphology**

✓ **Design of scratches on the surface can lead to complex surface structures due to multiple SPP-laser interference.**

Conclusions

1) **We identify two key components of LIPSS formation: SPP excitation and material-reorganization.**
2) **MD-TTM simulations [4,5] with the laser-SPP source term [6] are able to describe LIPSS formation.**
3) **A remarkable good agreement in LIPSS properties between experiment and simulations is found.**
4) **Manipulation of the laser fluence shape can pave a way for design of surface structures.**

References

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