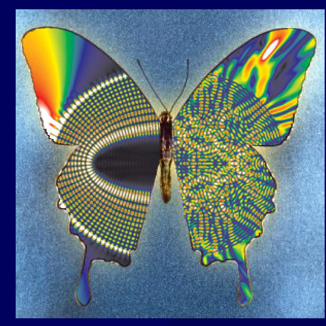
Growth of laser-induced surface nanostructures

Jean-Luc Déziel[1], Joey Dumont[1], Denis Gagnon[1], Louis J. Dubé[1], Sandra H. Messaddeq[2] and Younès Messaddeq[2]

[1]*Département de physique, de génie physique, et d'optique, Université Laval, Québec, Canada*
[2]*Centre d'optique, photonique et laser, Université Laval, Québec, Canada*



Nanostructured damage patterns

Strongly organized **laser-induced periodic surface structures** (LIPSSs) can be fabricated on **any type of solid** with intense femtosecond laser pulses, near the ablation threshold. LIPSSs allow us to:

- Tune material's optical properties (color, reflectivity, etc.),
- Make a surface super-hydrophopic,
- Study **intense light-matter interactions** on a **femtosecond time scale**.

The **Sipe-Drude theory** can predict the triggering of LIPSSs growth by solving the **analytical solutions** of Maxwell's equation for the propagation of an incident plane wave through a **random rough surface.**

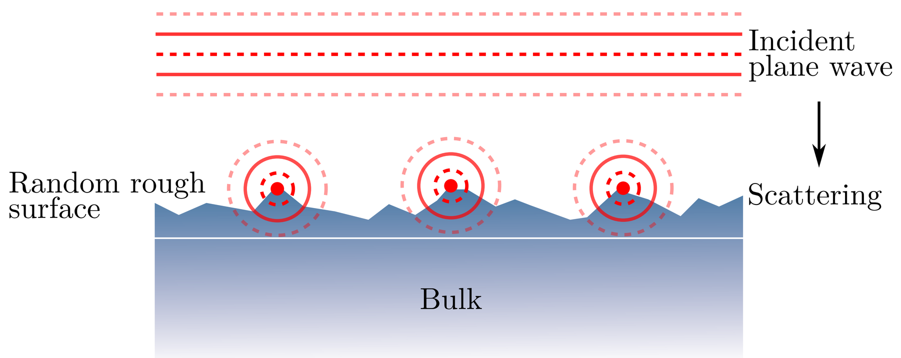


Fig. 1: Propagation and symmetry breaking of an incident plane wave.

Various LIPSSs morphologies can be predicted and fabricated depending on the laser properties **(wavelength λ, polarization)** and on the material's optical behavior **(dielectric, metallic).**

Dielectric behavior

Sipe-Drude theory predicts structures orientated **parallel** to the indicent light polarization for a material with **dielectric** optical properties ($\mathrm{Re}(\tilde{n}) > \mathrm{Im}(\tilde{n})$, where $\tilde{n}$ is the complex refractive index). The predicted LIPSSs periodicity Λ is equal to $\lambda/\mathrm{Re}(\tilde{n})$.

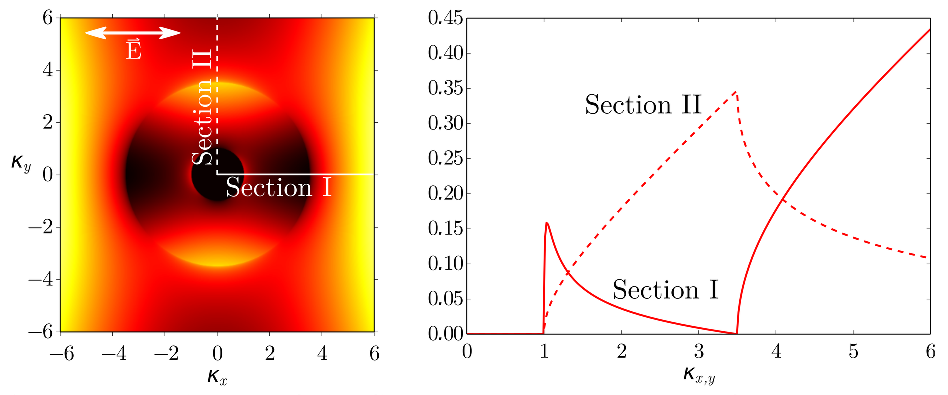


Fig. 2: Analytical solution of the Sipe-Drude theory in the frequency domain for a typical **dielectric** (silicon, $\tilde{n} = 3.5 + 0.0001i$).

Metallic behavior

For a strongly absorbing material, a **metallic** behavior ($\mathrm{Re}(\tilde{n}) < \mathrm{Im}(\tilde{n})$), structures orientated **perpendicular** to the incident light polarization are predicted. The predicted LIPSSs periodicity Λ is equal to λ.

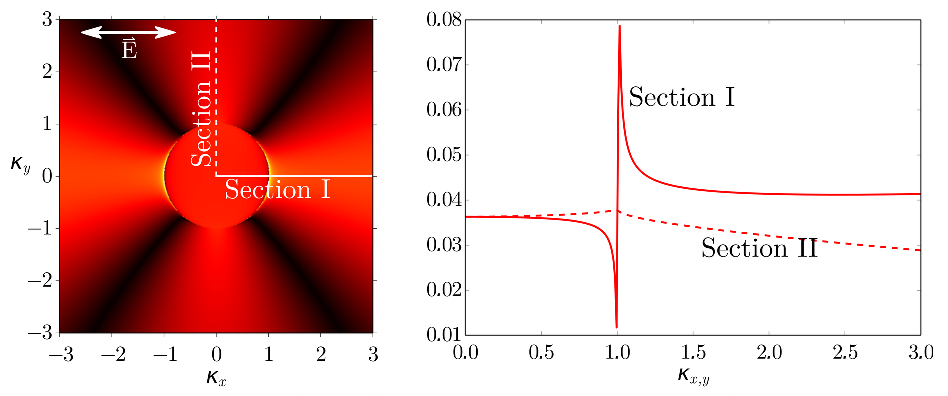


Fig. 3: Analytical solution of the Sipe-Drude theory in the frequency domain for a typical **metal** (aluminium, $\tilde{n} = 1.8 + 9.3i$).

Full wave simulations

The **finite-difference time-domain** (FDTD) method, while more involved than the Sipe-Drude analytical (but approximate) solution, provides deeper insights. For instance, the simulation results are:

- available in the **spatial domain**,
- available over all the simulation domain and the important **bulk** region,
- accurate for **high frequency** structures,
- easily **extensible** to account for other processes (feedback mechanism for instance).

The surface roughness is modelled by a **randomly generated binary function** over the material surface. Specifically, one random computational cell out of ten is filled with material.

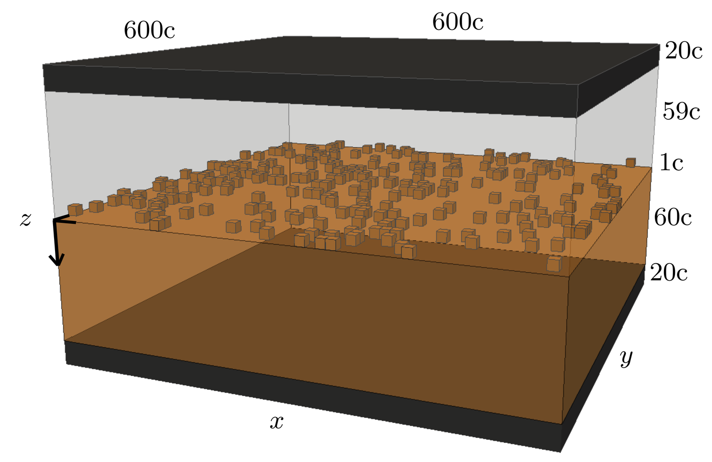


Fig. 4: Geometry used in the FDTD simulations. Dimensions are indicated in terms of numbers of computational cells.

The optical properties of the material are determined by the use of the **Drude model** as a function of the **plasma frequency**

$$\omega_p = \left[\frac{e^2 N_e}{\epsilon_0 m^*_{\mathrm{opt}} m_e}\right]^{1/2}, \qquad (1)$$

which depends on the free carriers density N_e. The dynamics of these free carriers is tuned with a second parameter, **the collision frequency γ**, defined as the inverse of the Drude damping time.

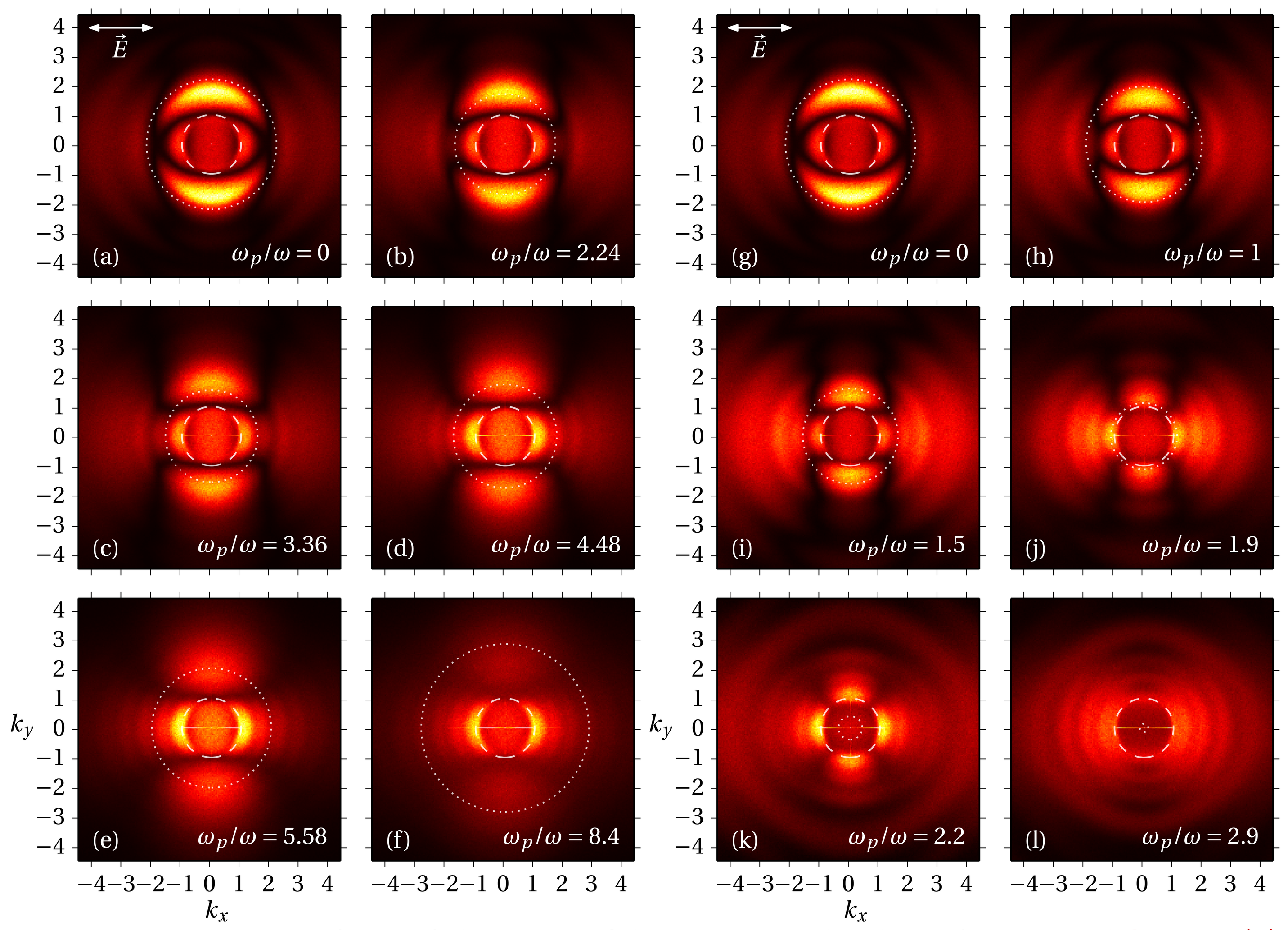


Fig. 5: Fourier transforms of the average field intensity under the surface. In **subfigures (a) to (f), $\gamma/\omega = 1$** and a transition from a dielectric to a metallic behavior is observed with the densification of the generated plasma. In **subfigures (g) to (l), $\gamma/\omega = 1/16$** and bidimensional structures, **crossed LIPSSs**, appear near the dielectric/metallic transition (see (k)).

Constructive & destructive feedback

Self-organization effects can be included with the addition of a **feedback mechanism**. This is achieved by **modifying the surface morphology** in the FDTD simulations according to the field spatial distribution **before sending another laser pulse**. By repeating this process for a number N of pulses, the surface morphology and the field distribution gradually build an equilibrium.

Growth on metals

To grow self-organized structures on a material with metallic behavior, the surface modification process has to be **ablation-like**, meaning that material is removed where the energy is maximum. The energy is maximum under the surface minima, with LIPSSs acting as **divergent lenses.**

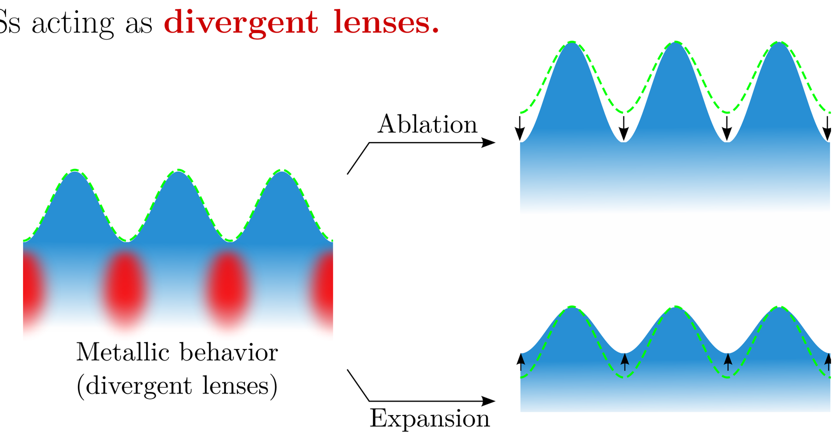


Fig. 6: Incident light is mostly directed to red spots. Amplitude growth (constructive feedback) on materials with metallic behavior is achieved by deeper ablation under the surface minima.

Growth on dielectrics

An **inverse mechanism** is used to grow LIPSSs on materials with dielectric behavior, **photo-expansion**. In this case, material is added where the energy is larger, over the surface maxima acting as **convergent lenses** in this case.

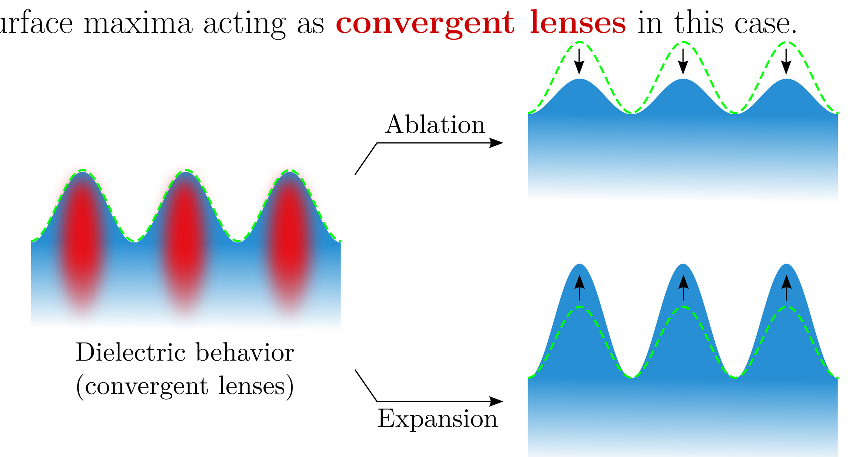


Fig. 7: Incident light is mostly directed to red spots. Amplitude growth (constructive feedback) on materials with dielectric behavior is achieved by expanding surface maxima.

Self-organization and growth

An ablation-like feedback mechanism is implemented to grow structures orientated perpendicular to the light polarization on a material with metallic behavior. **Every parts of the bulk which receive more light intensity than a certain threshold is removed between each laser pulse.**

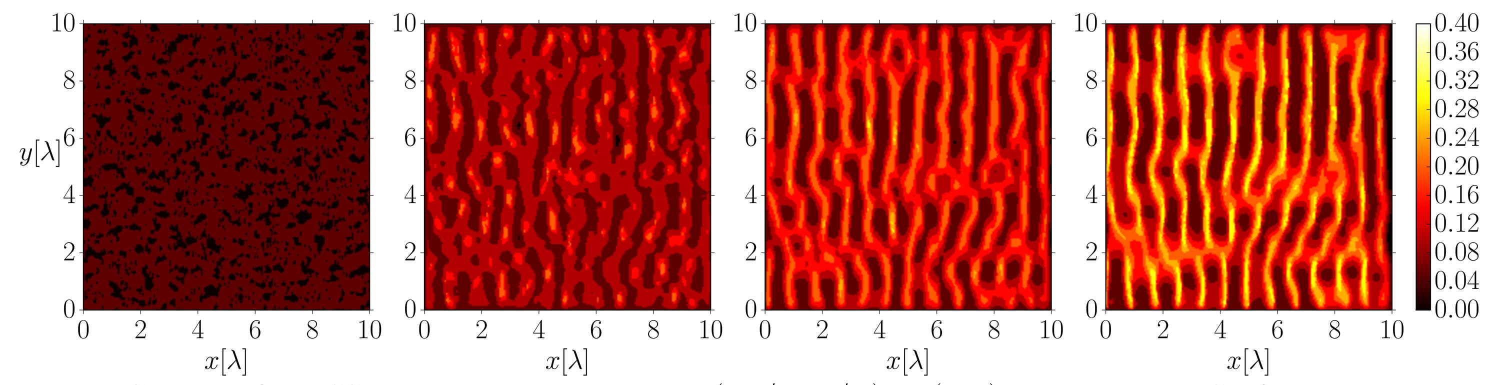


Fig. 8: Growth of LIPSSs on a Drude metal with $(\omega_p/\omega, \gamma/\omega) = (8, 1)$ and ablation. Surface morphology is shown for pulse numbers $1, 3, 5, 8$.

To grow structures orientated parallel to the light polarization on dielectrics, an **expansion-like** feedback mechanism is used and implemented with the same threshold method as for ablation, except the **surface position modification is applied with an opposite sign.**

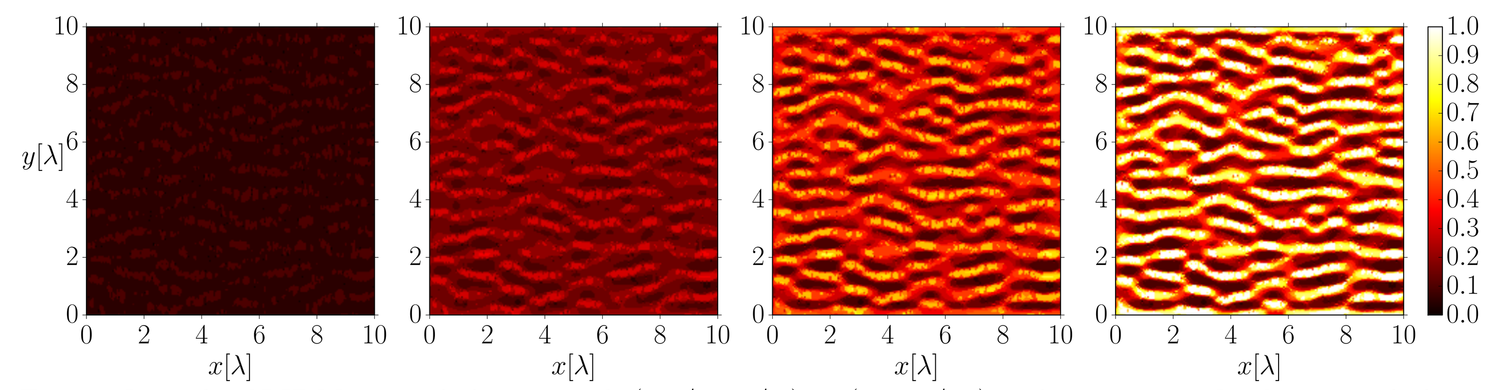


Fig. 9: Growth of LIPSSs on a dielectric with $(\omega_p/\omega, \gamma/\omega) = (1.7, 1/16)$ and expansion. Surface morphology is shown for pulse numbers $1, 2, 3, 4$.

Ackowledgments

The authors acknowledge the financial support form the Canada Excellence Research Chair in Photonics Innovations of Y. Messaddeq and the Natural Sciences and Engineering Research Council of Canada (NSERC). We also acknowledge computational resources from Calcul Québec and the free software project Meep.

NSERC CRSNG

Calcul Québec

References

Full wave simulations and crossed LIPSSs are discussed in:
1. J. -L. Déziel, J. Dumont, D. Gagnon, L. J. Dubé, S. H. Messaddeq, and Y. Messaddeq, arXiv:1410.0583v2 (*to be published in Journal of Optics*)

Derivation of Sipe's analytical solutions are in:
2. J. E. Sipe, J. F. Young, J. S. Preston, and H. M. van Driel, Phys. Rev. B 27, 1141 (1983).

More on feedback applied to LIPSSs growth:
3. J. Z. P. Skolski, G. R. B. E. Römer, J. V. Obona, and A. J. Huis in't Veld, J. Appl. Phys. 115, 103102 (2014).