

Laboratoire Hubert Curien

Saint-Etienne, France



**Plasmonic and hydrodynamic effects
in ultrafast laser generation of
periodic surface structures
on metals**

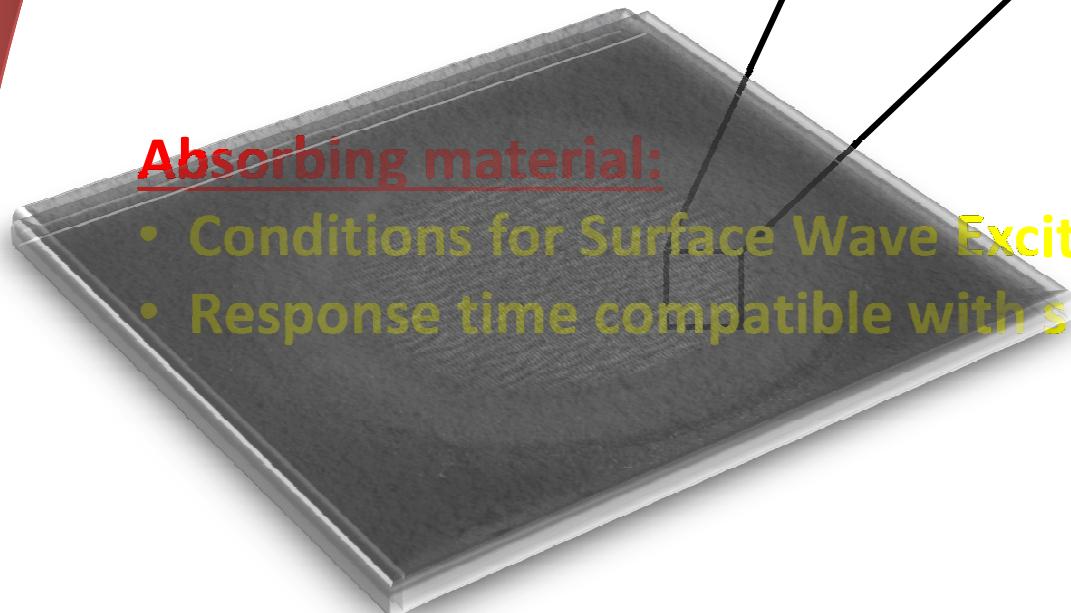
J.P. Colombier

Formation dynamics of LIPSS



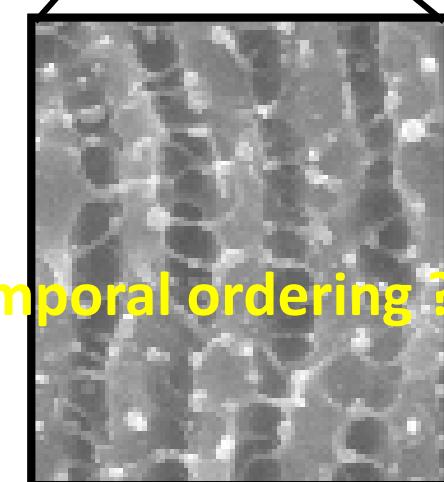
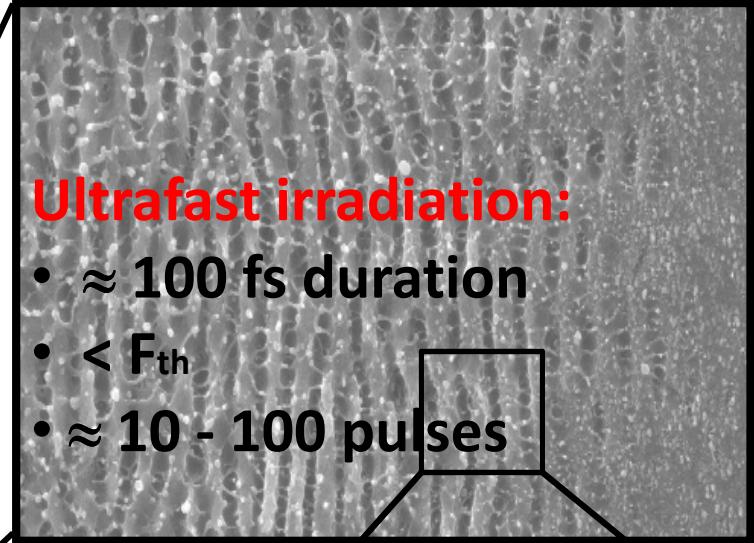
Low spatial frequency LIPSS:

- Perpendicular to E
- $\approx 600 \text{ nm period}$ (*several shots*)

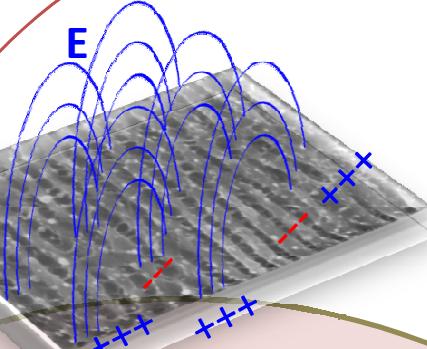


Absorbing material:

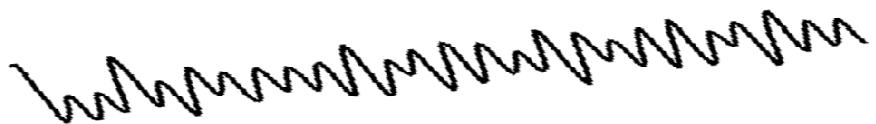
- Conditions for Surface Wave Excitation ?
- Response time compatible with spatio-temporal ordering ?



Multiscale structuration



Origin : 2 main concepts



Interference model and/or plasmon waves excitation

Resonant in nature,
inhomogeneous energy
deposition...

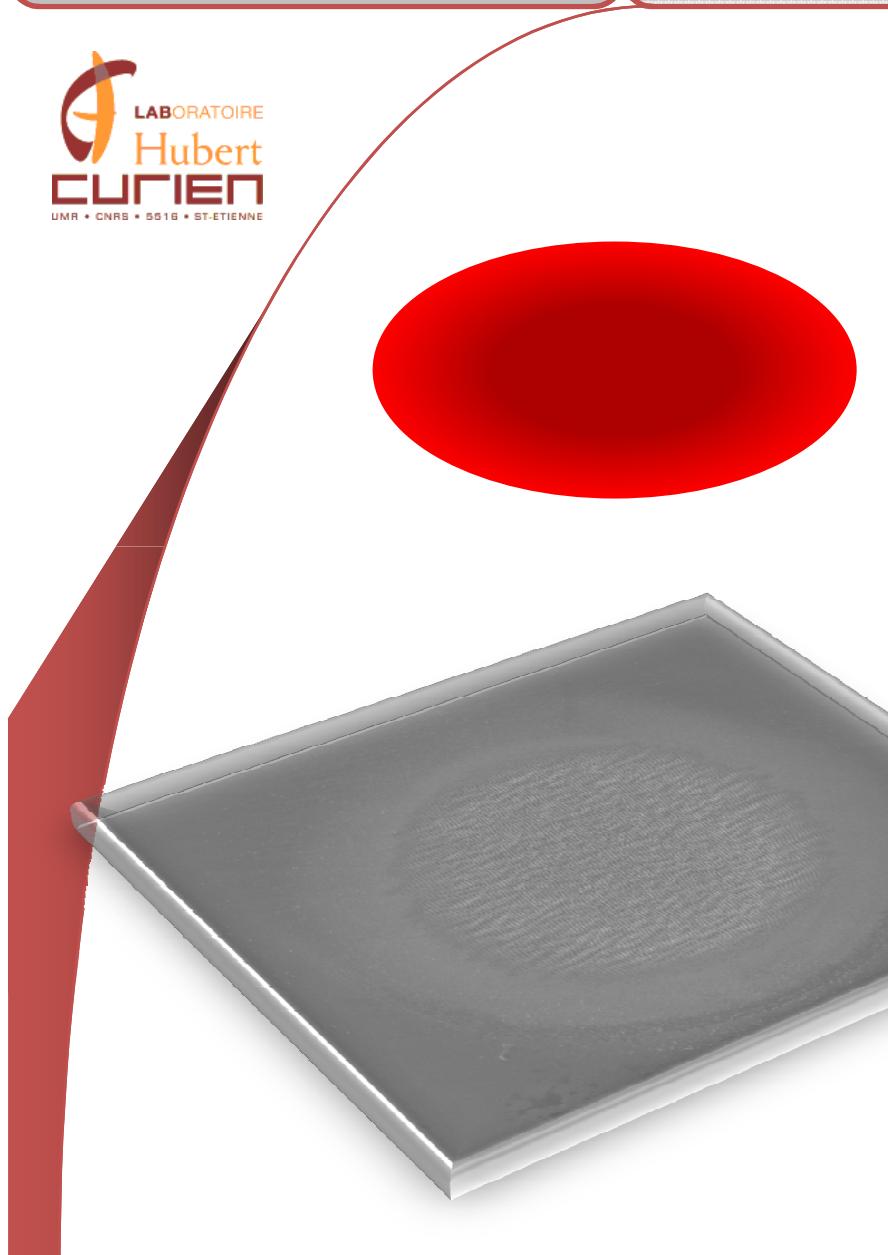
- ⇒ Agree with the most observed orientation & period
- ⇒ Dependence on laser fluence and pulse number ?

Spatio-temporal ordering: spontaneous pattern formation

Instability, positive feedback and exponential growth of initial perturbation...

- ⇒ Agree with periodicity dependence on Fluence/N
- ⇒ Orientation and laser wavelength dependence ?

Hybrid nature ⇒ Towards a unified approach ?



Step by step formation

Overview:

- **Absorption stage**
Plasmonic experiment
- **Nonequilibrium energy distribution**
LIPSS contrast on different materials
- **Hydrodynamic stage**
2T simulation
- **Positive feedback effects**
Discussion on characteristic timescales

Grating-coupled SP experiment

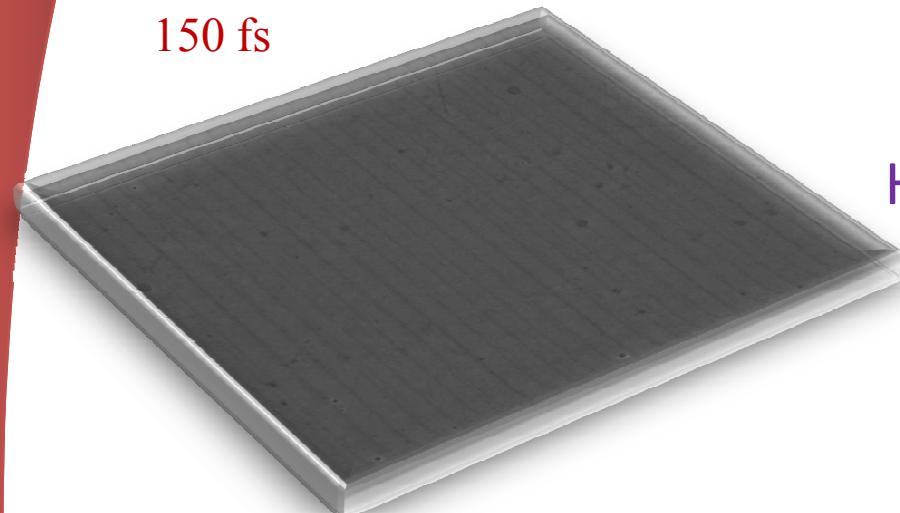


Basic idea: A photon cannot transform into a SP without some form of momentum conservation

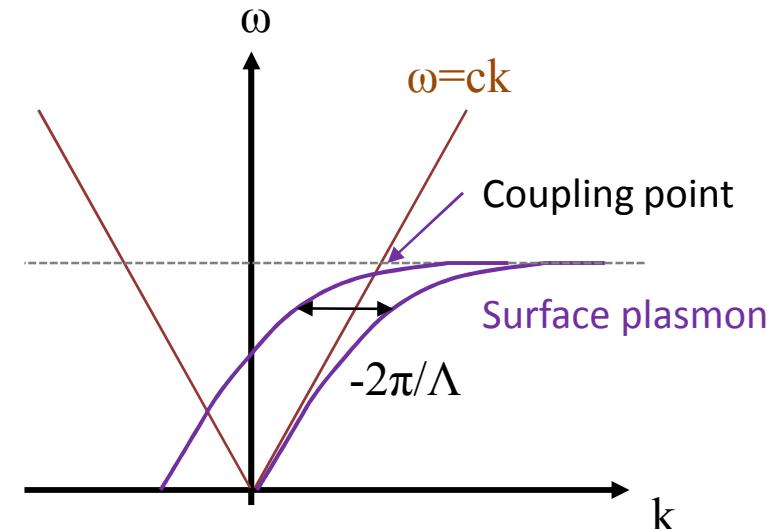
Rough surface \approx superposition of different gratings

Single shot

$\lambda=800 \text{ nm}$
 150 fs



$$k_{\text{SP}} = k_t \pm n q_g$$



How to correlate SP and ripples ?

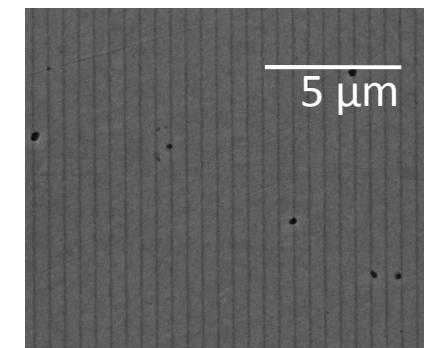
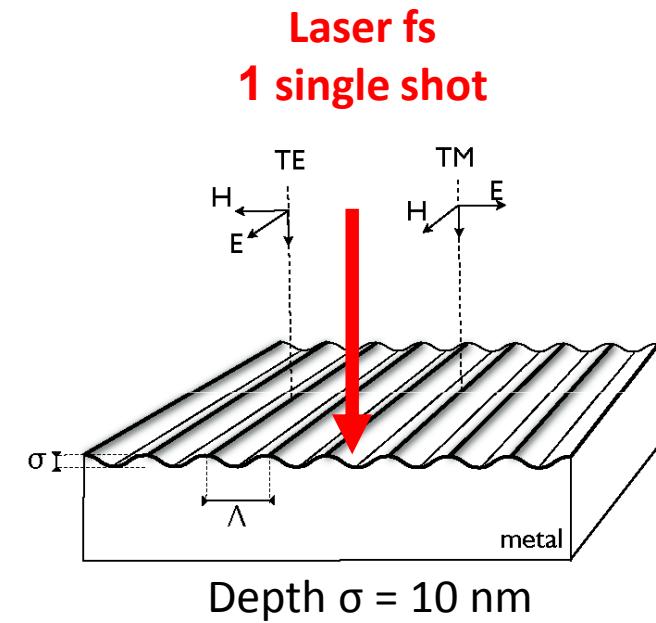
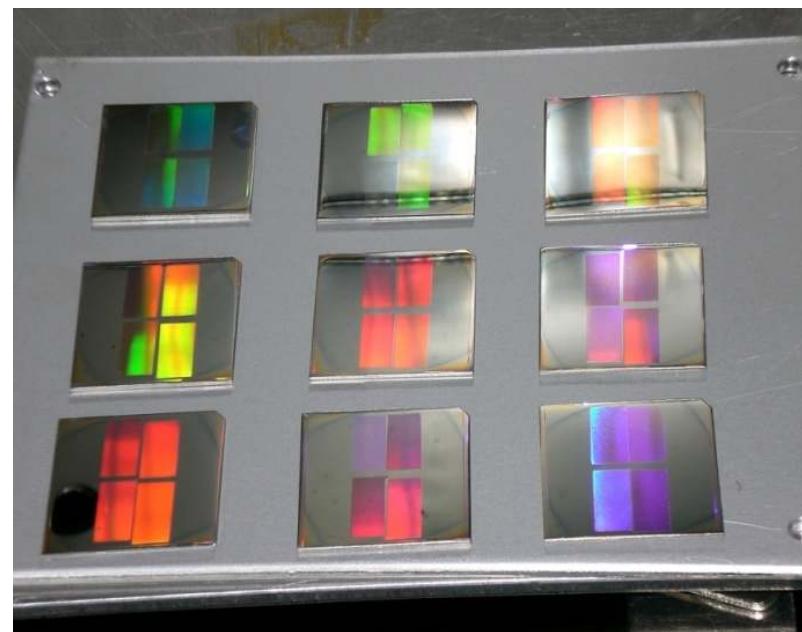
⇒ Find resonance for a set of gratings with controlled Λ

Grating-coupled SP experiment



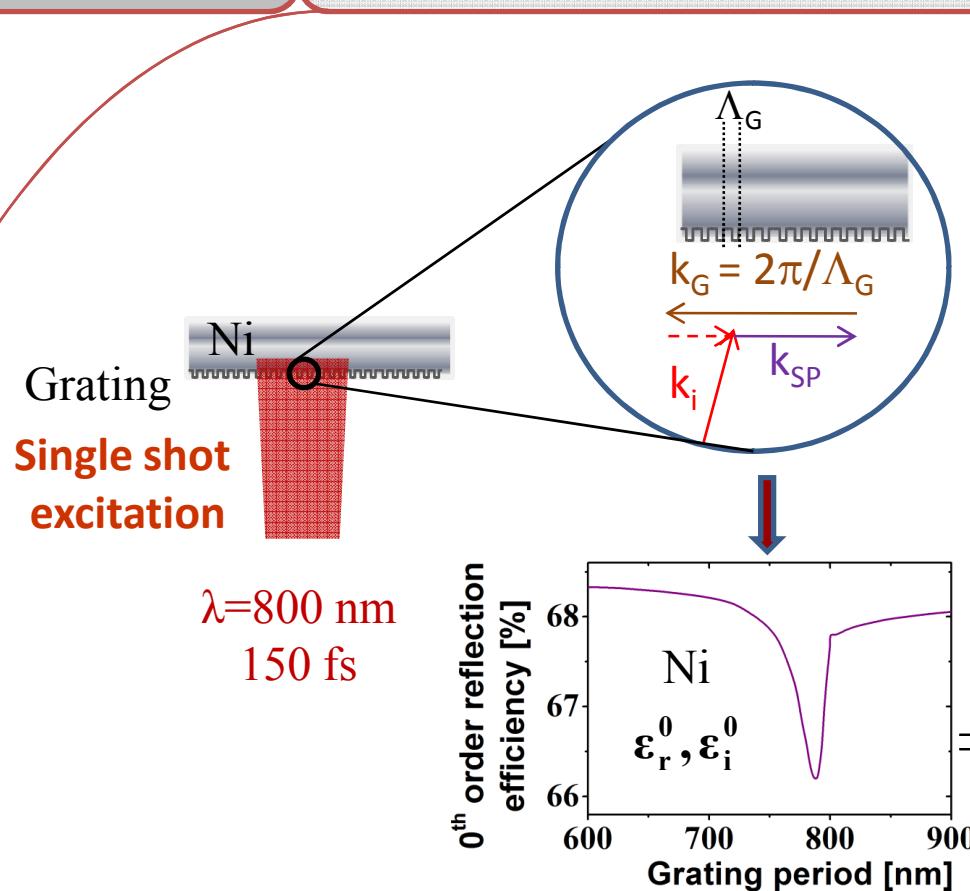
Mach-Zehnder interference scheme

Grating periods on Nickel range
from $\Lambda_G = 440 \text{ nm}$ to $\Lambda_G = 800 \text{ nm}$
with 10 nm increment



F. Garrelie et al., Opt. Express, 19 (10), 9035 (2011)

Grating-coupled SP experiment



Λ_{LIPSS} : Ripples periodicity

Λ_G : Grating periodicity

$$\frac{1}{\Lambda_{\text{LIPSS}}} = \frac{\sin \theta}{\lambda} \pm \frac{N}{\Lambda_G}$$

$\theta=0$: Normal incidence
 1^{st} diffraction order

⇒ Larger coupling efficiency

$$\Lambda_{\text{LIPSS}} = \Lambda_G$$

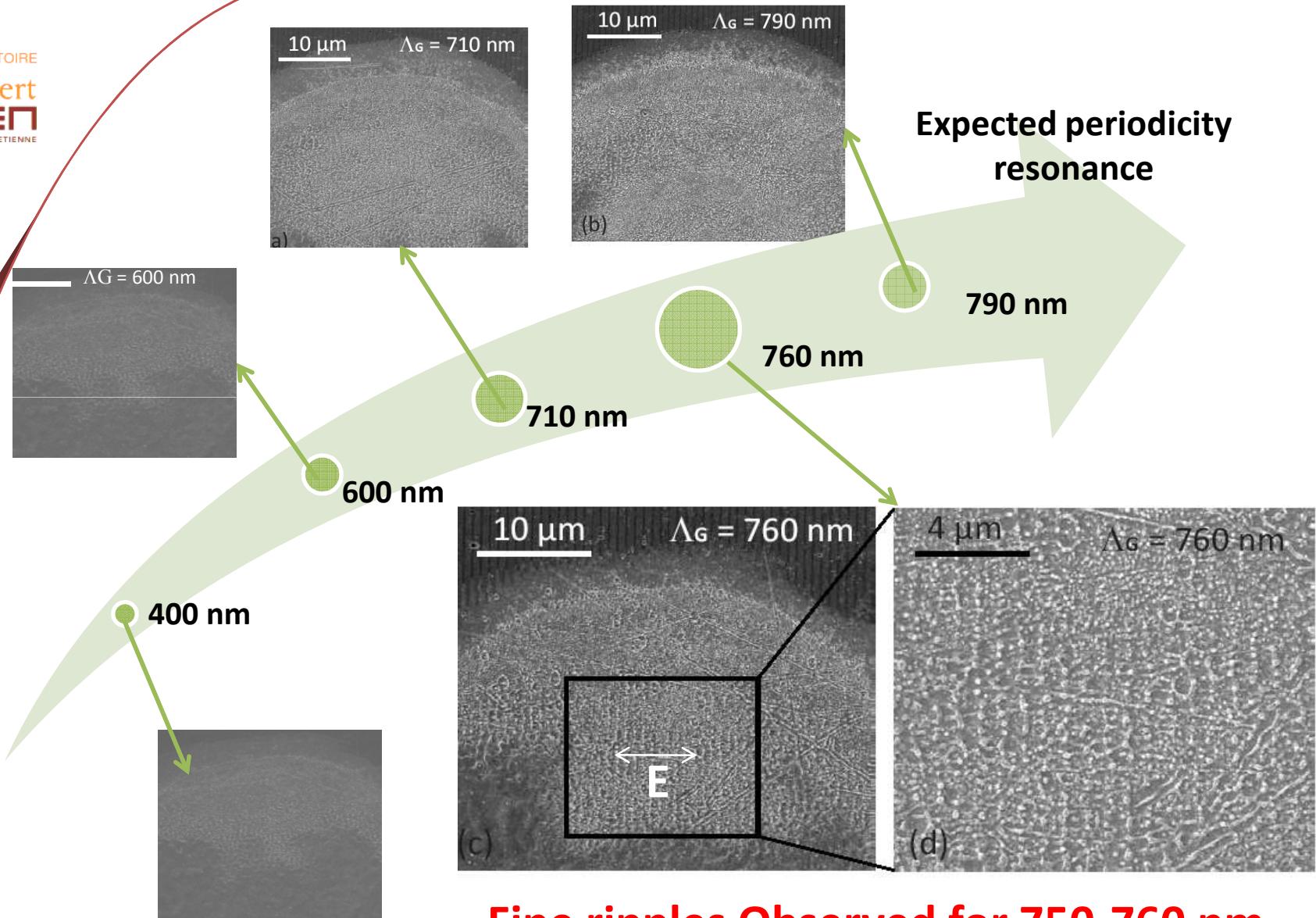
Surface Plasmon excitation

⇒ Ripples formation for a well-defined Λ_G & TM irradiation only

$$\Lambda_{\text{LIPSS}} = \Lambda_G = \lambda_{\text{SP}} = \frac{2\pi}{k'_{\text{SP}}} = \lambda \Re \left[\left(\frac{\epsilon_d + \tilde{\epsilon}_m}{\epsilon_d \tilde{\epsilon}_m} \right)^{1/2} \right] = \underline{\underline{790 \text{ nm}}} \\ \text{for Nickel @ 800 nm}$$

*Plasmonic & Hydro
formation of fs LIPSS*

Irradiation results

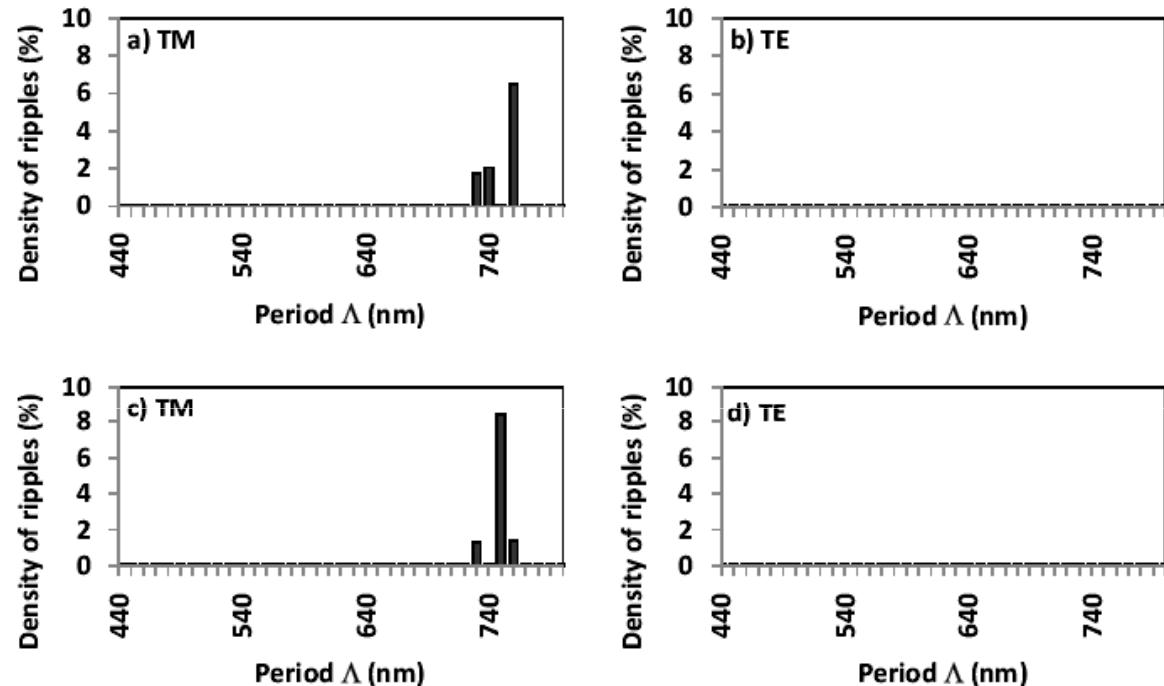


Analysis

$F = 0.97 \text{ J/cm}^2$

$F = 1.42 \text{ J/cm}^2$

Density of ripples as a function of the grating period for 2 irradiation fluences

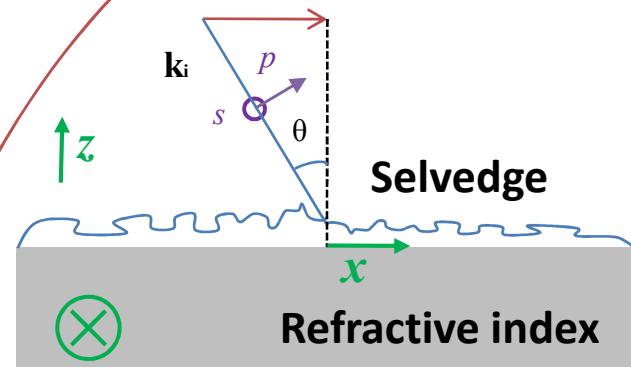


*Ratio between surface covered
with fine ripples and the overall surface of the spot*

SP resonance : Ripples formation for
✓ TM irradiation
✓ and $\Lambda_G = 750\text{-}760 \text{ nm}$

*Plasmonic & Hydro
formation of fs LIPSS*

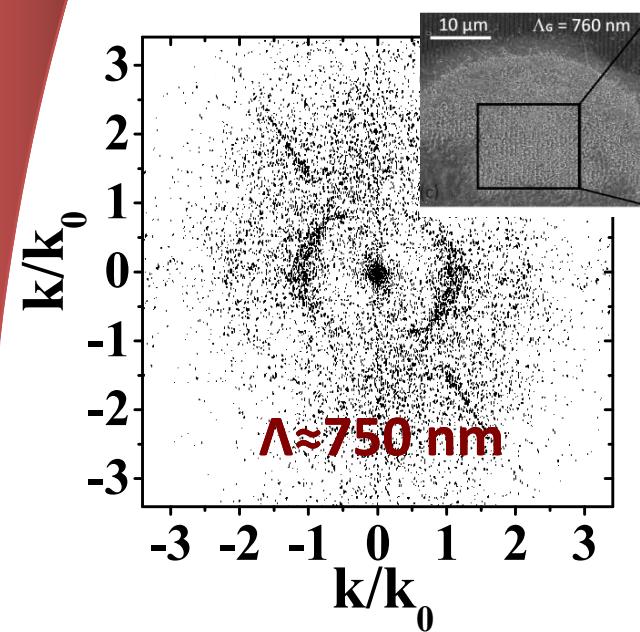
Sipe calculation



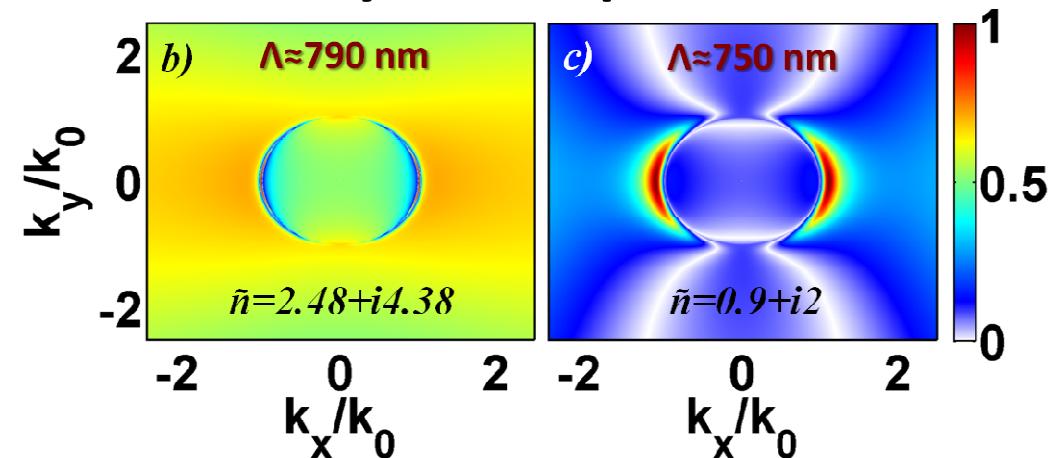
« Surface-scattered » fields interfere
with the incident beam

Inhomogeneous absorption mainly determined
by an **efficacy η** depending upon k

J. E. Sipe, J. F. Young, J. S. Preston and H. M. van Driel, Phys. Rev. B 27, 1141 (1983).



Efficacy factor η calculation



Transient change of optical properties ?

Calculating plasmon wavelength

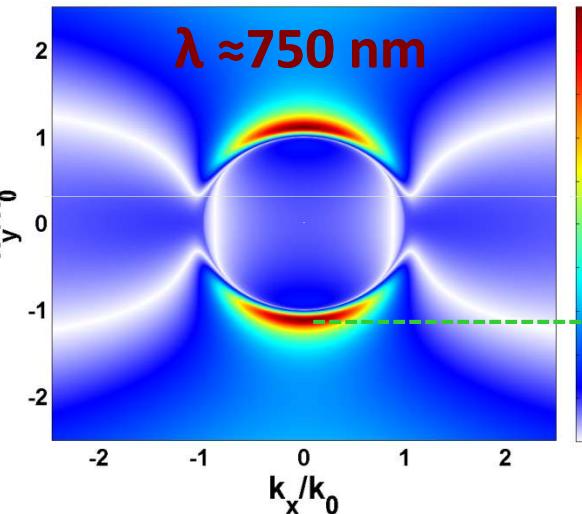


Theoretical plasmon wavelength

Sipe model

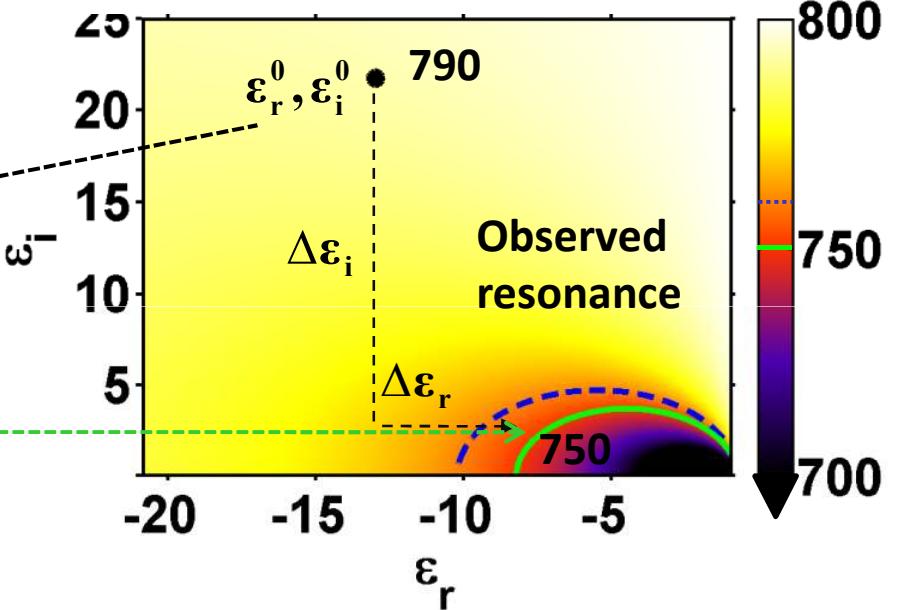
Efficacy factor

$\lambda \approx 750 \text{ nm}$



Inhomogeneous energy deposition

$$\lambda_{SP} = \frac{2\pi}{k'_{SP}} = \lambda \Re \left[\left(\frac{\epsilon_d + \tilde{\epsilon}_m}{\epsilon_d \tilde{\epsilon}_m} \right)^{1/2} \right]$$



Decrease of ϵ_i and $|\epsilon_r|$
required to explain observed
resonance

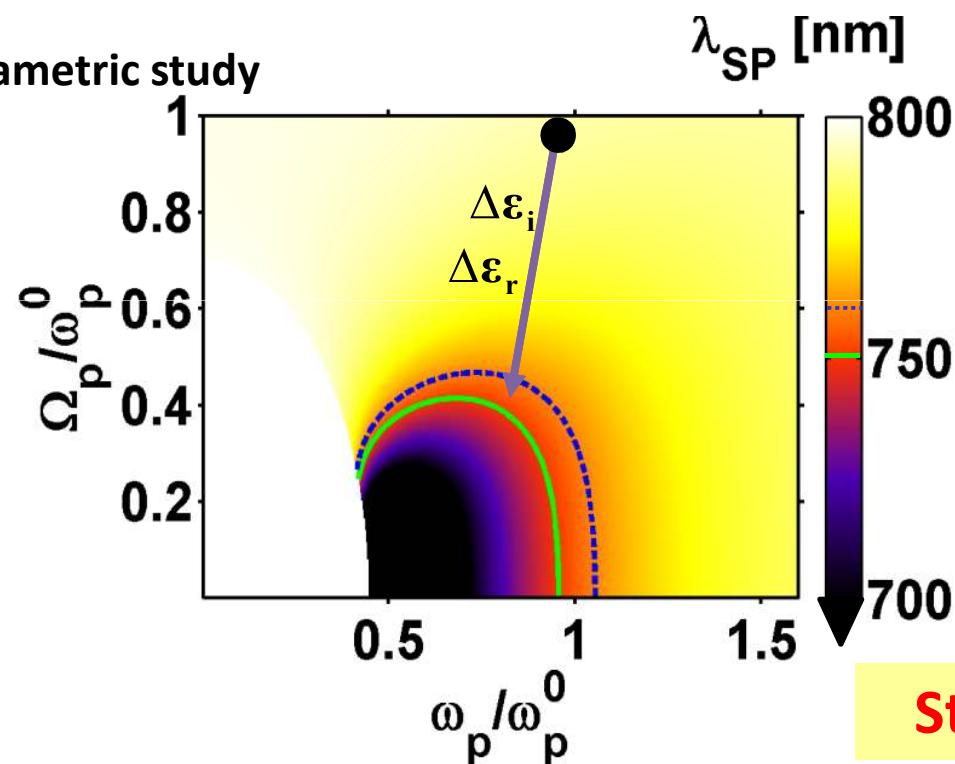
Calculating plasmon wavelength



DL model

$$\tilde{\epsilon}_m = \left[1 - \frac{f_0 \omega_p^2}{\omega(\omega - i\gamma)} \right]_D + \left[\sum_{j=1}^k \frac{f_j \Omega_p^2}{(\omega_j^2 - \omega^2) + i\omega\Gamma_j} \right]_{IB}$$

Parametric study



$$\left. \begin{array}{l} \omega_p^2 = \frac{n_1 e^2}{m_1 \epsilon_0} \\ \Omega_p^2 = \frac{n_2 e^2}{m_2 \epsilon_0} \end{array} \right\}$$

Depend on electronic
band structure
of the irradiated metal

Strong decrease of Ω_p

Decrease of the d-band electrons contribution
available to undergo an interband transition



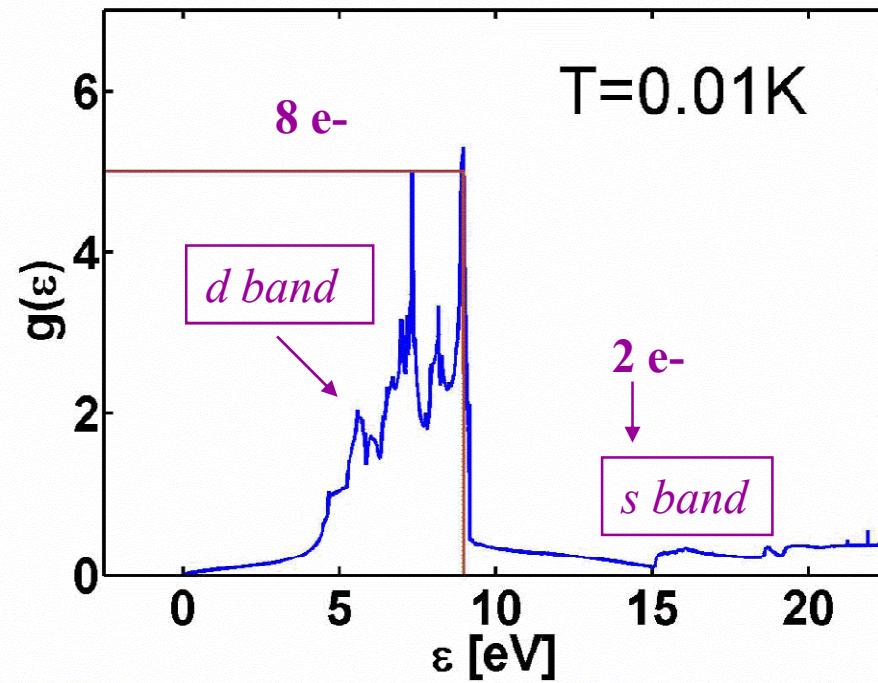
NICKEL

Low temperature:
 E_F falls in the *d*-band

High temperature:
Thermal excitation shifts the
chemical potential away from the
(high density) *d* band

⇒ Te increase modifies Ω_p ?

DOS [States/eV/atom]



Decrease of the *d*-band electrons contribution
available to undergo an interband transition ?

Questions on absorption stage



SP excitation

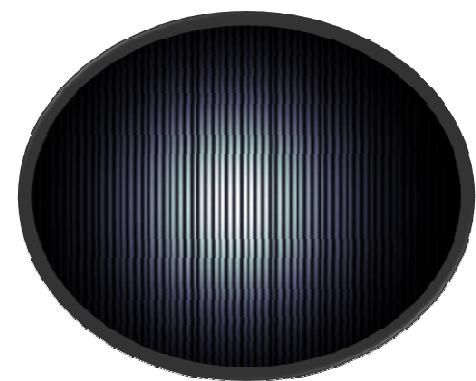
Direct damping of SP ?
Interference effect ?

**Spatially modulated heating
of the sample surface**

*Incident electric
field (laser)*
+
*Scattered surface
wave (SP)*

Interference intensity

$$I = I_0 + I_1 \cos(k_s x)$$

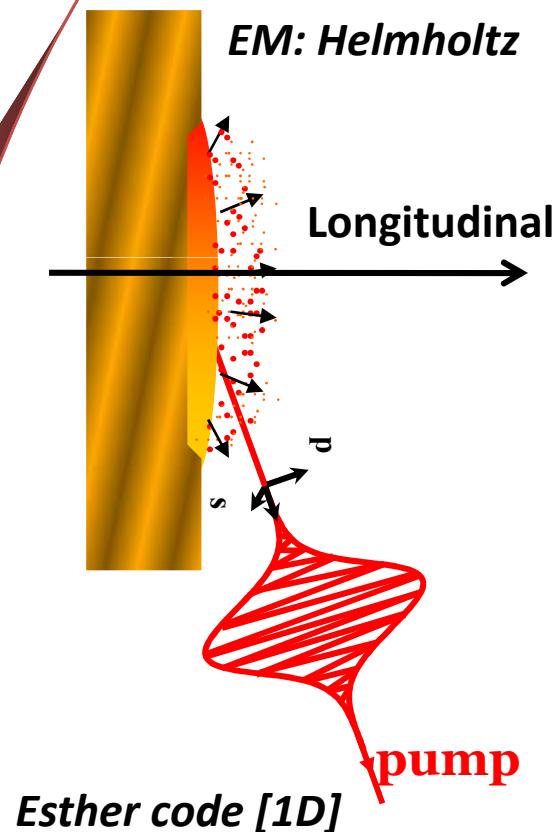


Parameters/events defining contrast ?
Spatio-temporal ordering ?

Gaussian distribution

*Plasmonic & Hydro
formation of fs LIPSS*

Hydrodynamic model



e-ph Coupling

Diffusion

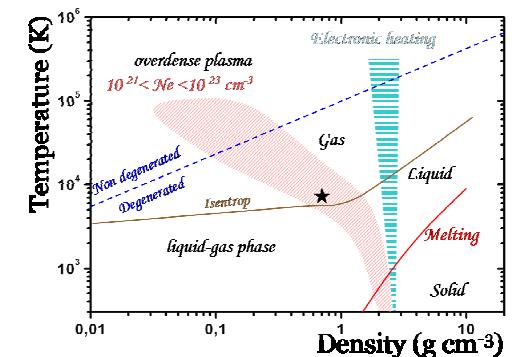
$$\frac{\partial \rho \epsilon_e}{\partial t} = \frac{\partial}{\partial z} \left(K_e \frac{\partial T_e}{\partial z} \right) - \gamma(T_e - T_i) - p_e \frac{\partial V}{\partial t} + \frac{n_e \xi_e}{Z^*} \frac{\partial Z^*}{\partial t} + \Re[\tilde{\sigma}] \frac{\tilde{E} \tilde{E}^*}{2}$$

$$\frac{\partial \rho \epsilon_i}{\partial t} = \frac{\partial}{\partial z} \left(K_i \frac{\partial T_i}{\partial z} \right) + \gamma(T_e - T_i) - p_i \frac{\partial V}{\partial t} - \frac{n_e \xi_e}{Z^*} \frac{\partial Z^*}{\partial t}$$

$$\frac{\partial u}{\partial t} = -\frac{\partial}{\partial m} (p_e + p_i), \quad \frac{\partial V}{\partial t} = \frac{\partial u}{\partial m},$$

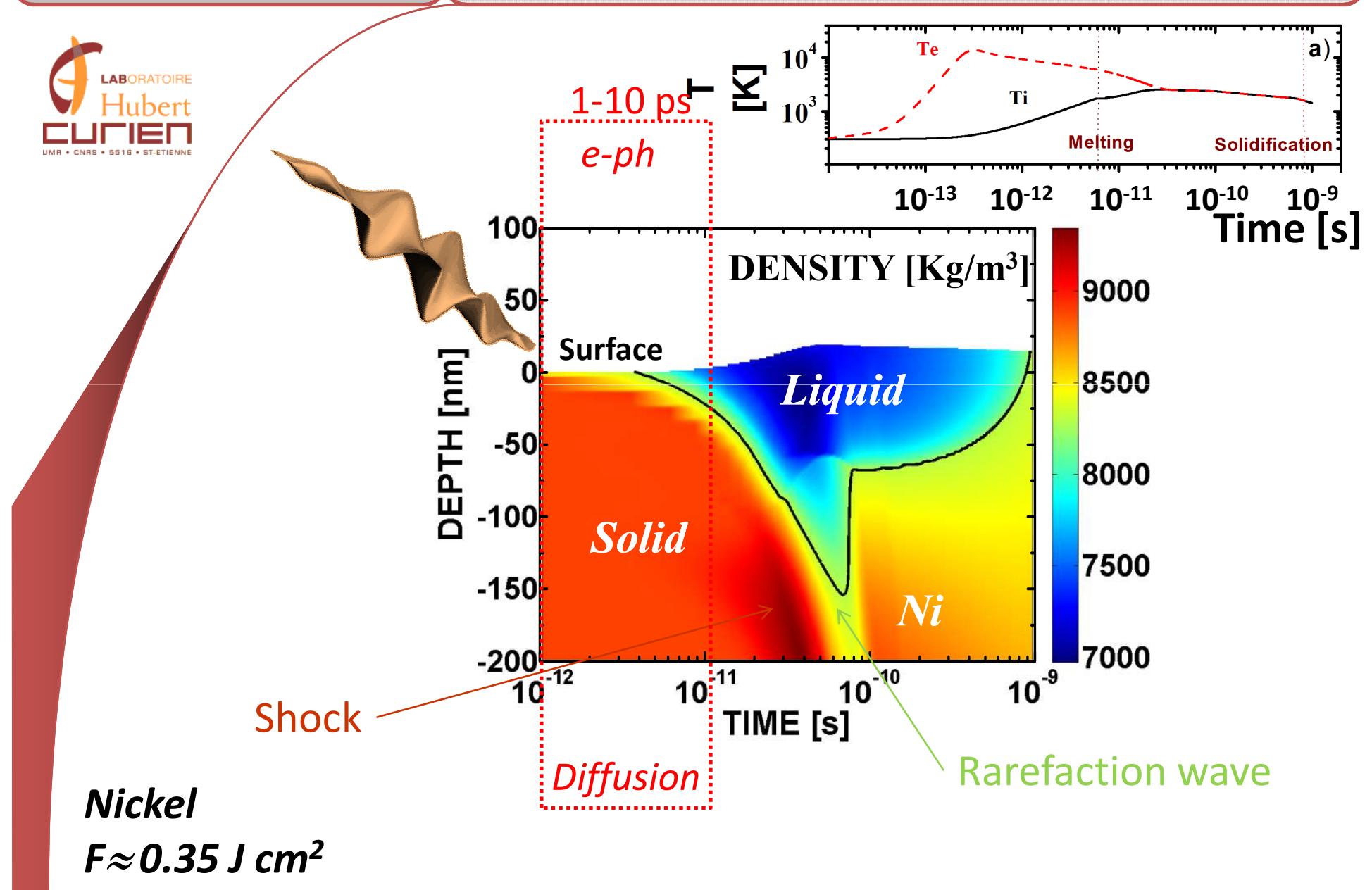
Hydro

Equation of states
[electrons (Thomas-Fermi)
+ ions (EOS BLF)]



*Plasmonic & Hydro
formation of fs LIPSS*

Phase transition



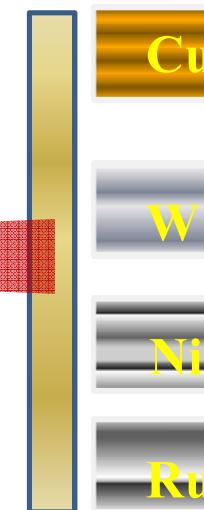
(*e-ph*) coupling experiment

Metals with different γ and behavior with Te

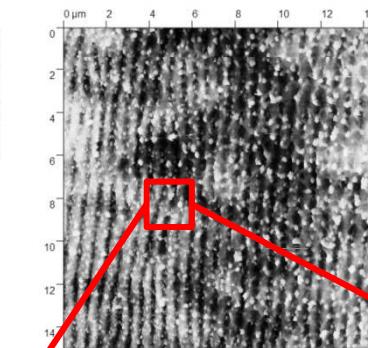
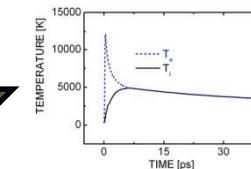
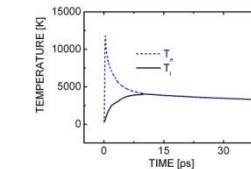
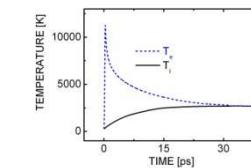
$\lambda=800$ nm

150 fs

50 pulses



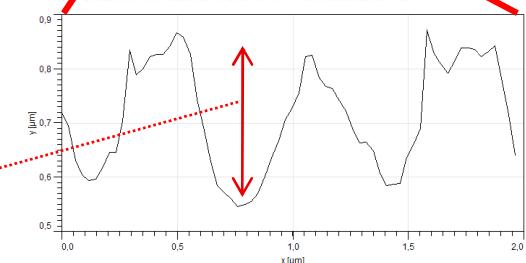
TTM



AFM

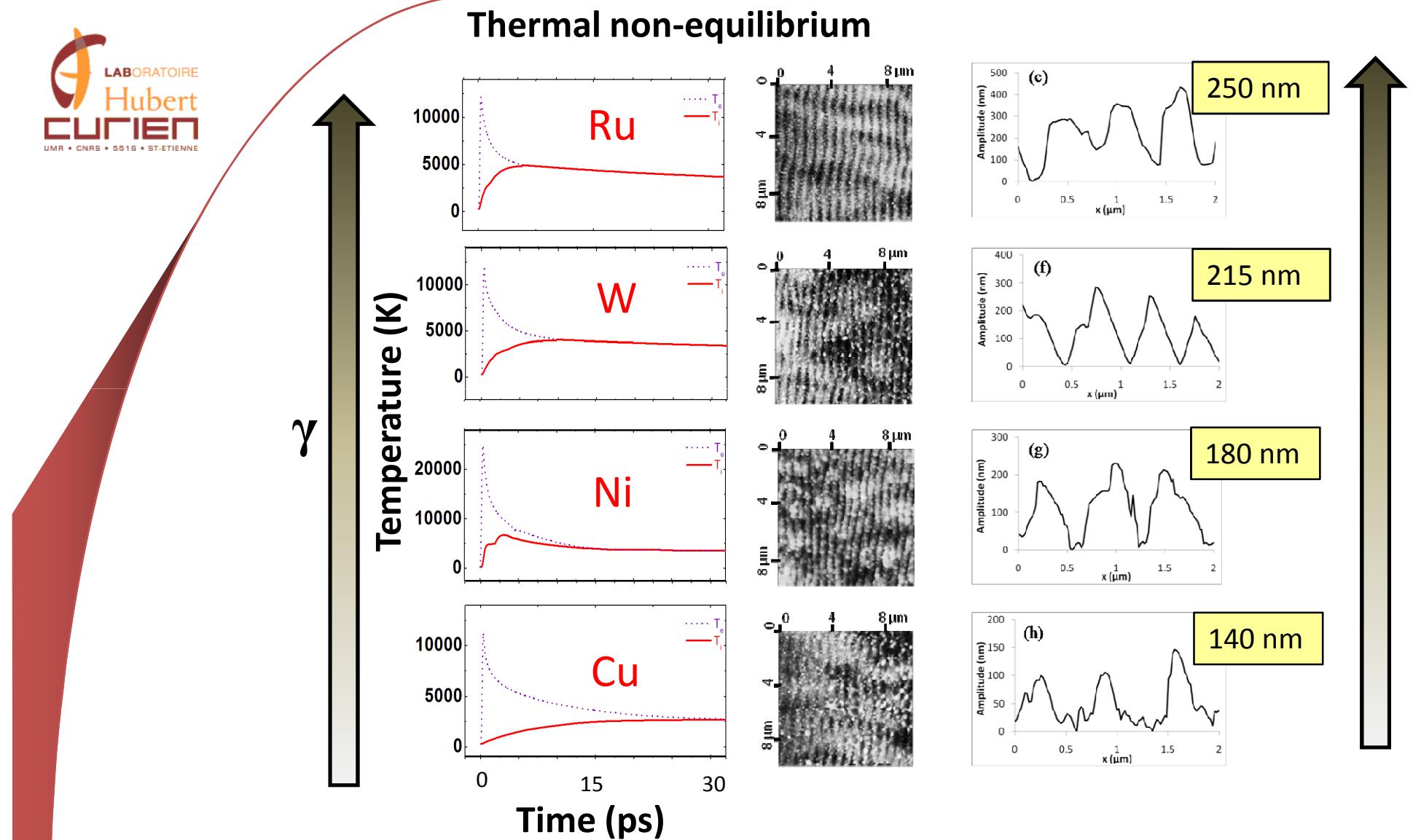
3 transition metals : Ni, W, Ru
1 Noble metal : Cu

Amplitude
measurement for
each metal



*Plasmonic & Hydro
formation of fs LIPSS*

(*e-ph*) coupling experiment

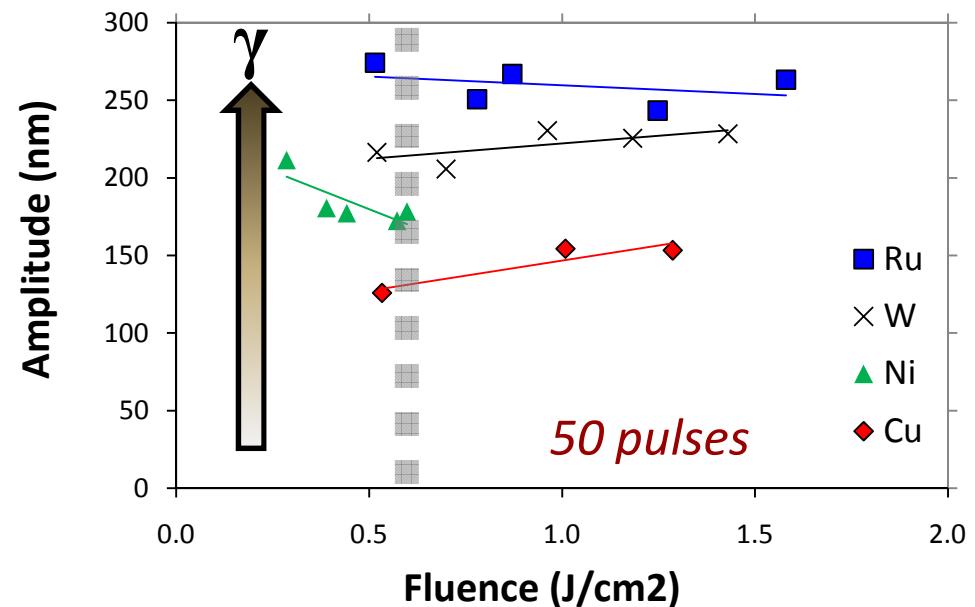


Evolution with fluence



Height of the cross-section of the surface ripples for Ru, W, Ni & Cu as a function of local fluence

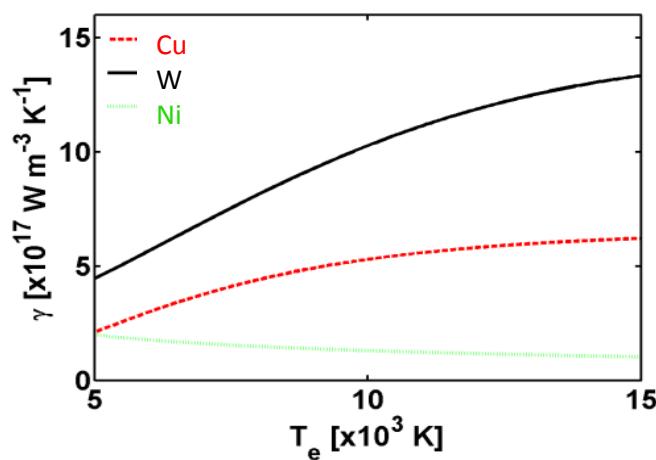
$A_R(\text{Ru})$ \rightarrow
 $A_R(\text{W})$ \nearrow
 $A_R(\text{Ni})$ \searrow
 $A_R(\text{Cu})$ \rightarrow



J.P. Colombier et al., J. Appl. Phys. 111, 024902 (2012)

γ evolution with T_e

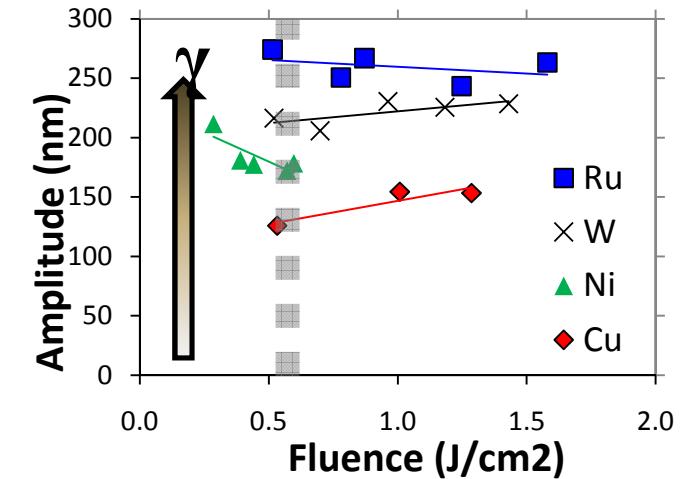
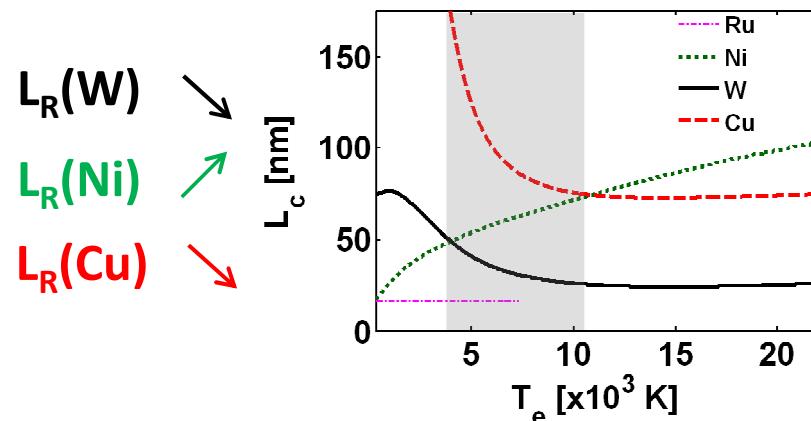
Lin et al., Phys. Rev. B 77 (2008)





*Correlation energy confinement ($\approx L_R$)
& ripples contrast ?*

Estimation of the confinement size
Electronic diffusion length L_R
 L_R dependence on $T_e \leftrightarrow F$
 [P. Corkum et al., PRL, 61, 2886 (1988)]

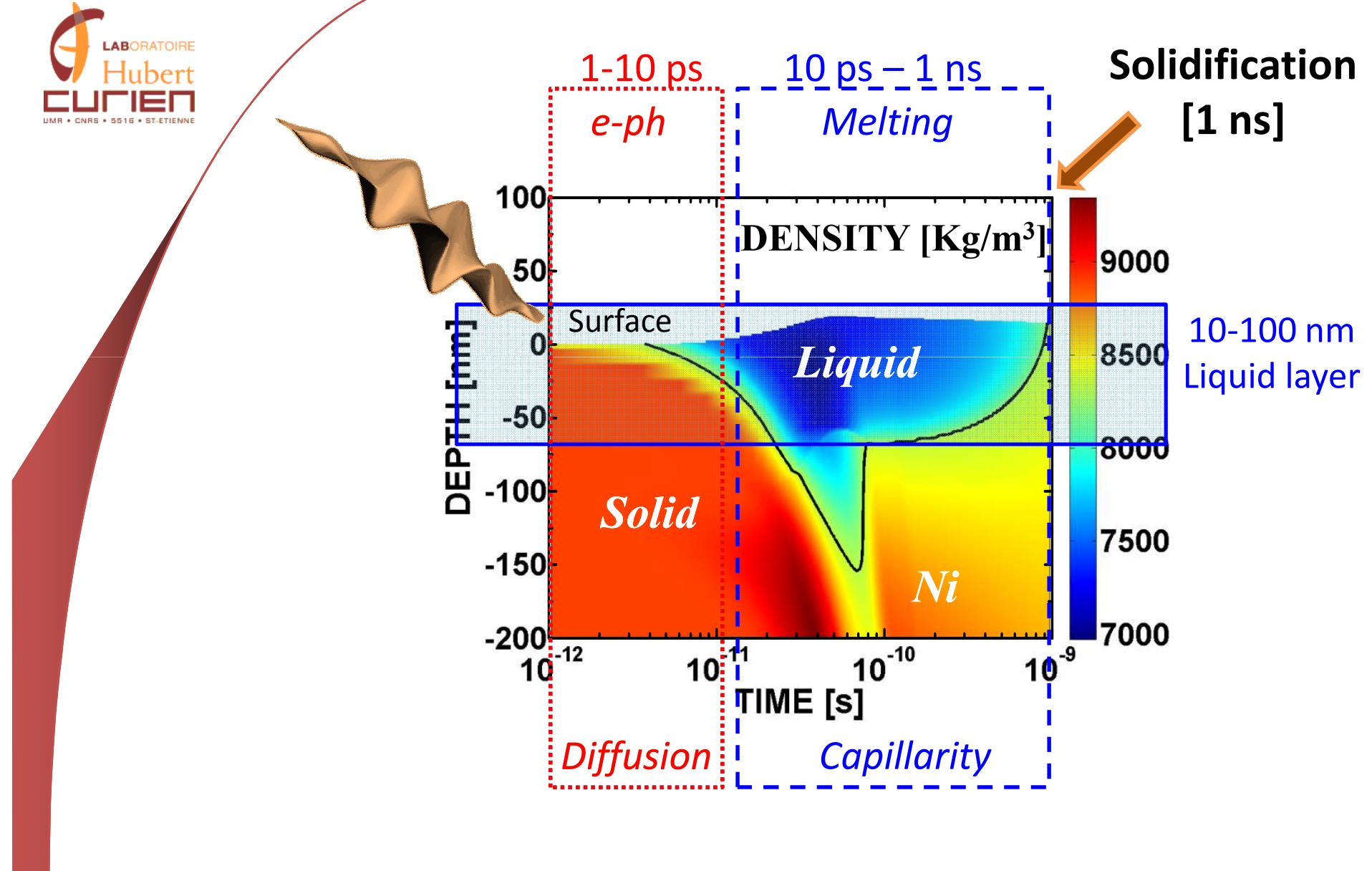


$$L_R = \left(\frac{128}{\pi} \right)^{1/8} \left(\frac{C_i}{A_e T_m} \right)^{1/4} \left(\frac{K}{\gamma} \right)^{1/2}$$

High energy confinement
 → **High ripples contrast**

*Plasmonic & Hydro
formation of fs LIPSS*

Feedback process



Feedback process (*Hydro*)

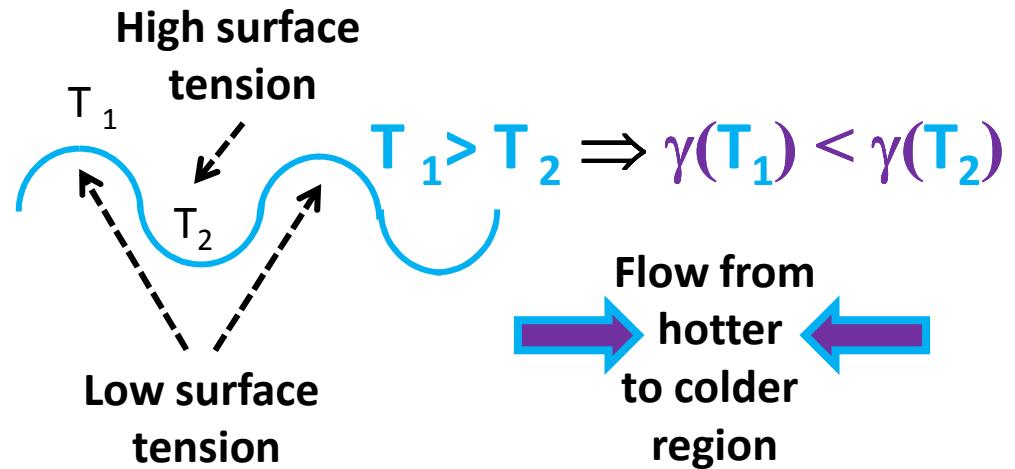


Surface tension of the molten surface upon heating may force to minimize the surface area



$$\nabla T \text{ causes } \nabla \gamma$$

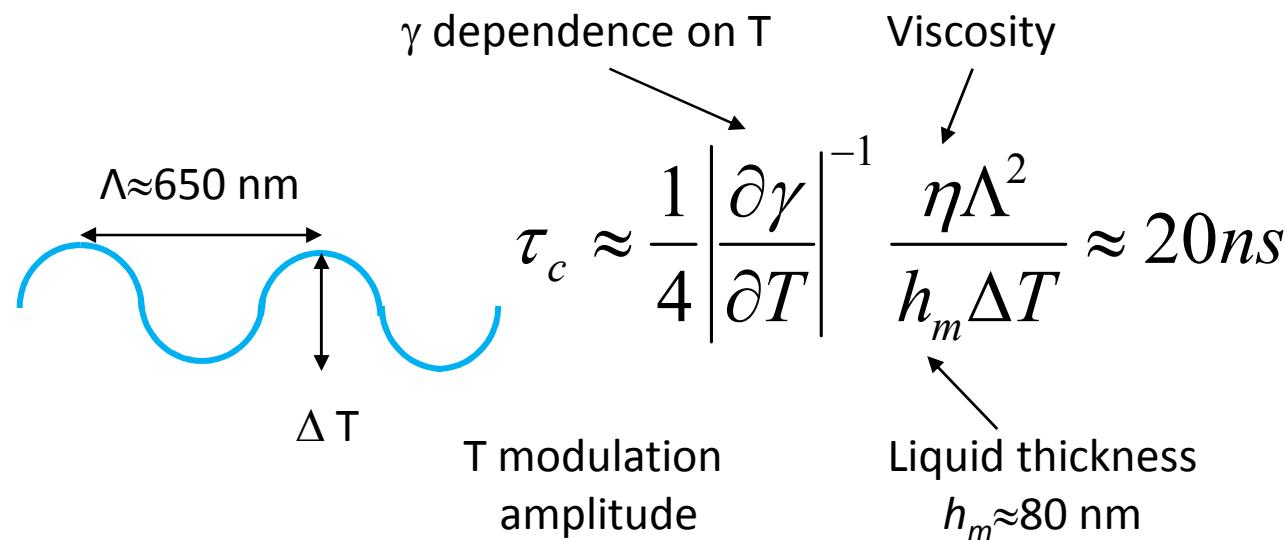
Thermocapillary force (Marangoni)



Evaluating the Marangoni timescale:

Transverse velocity of a laminar
liquid displacement

$$v_c \approx \frac{h_m \gamma_T}{\eta} \frac{\partial T}{\partial x}$$



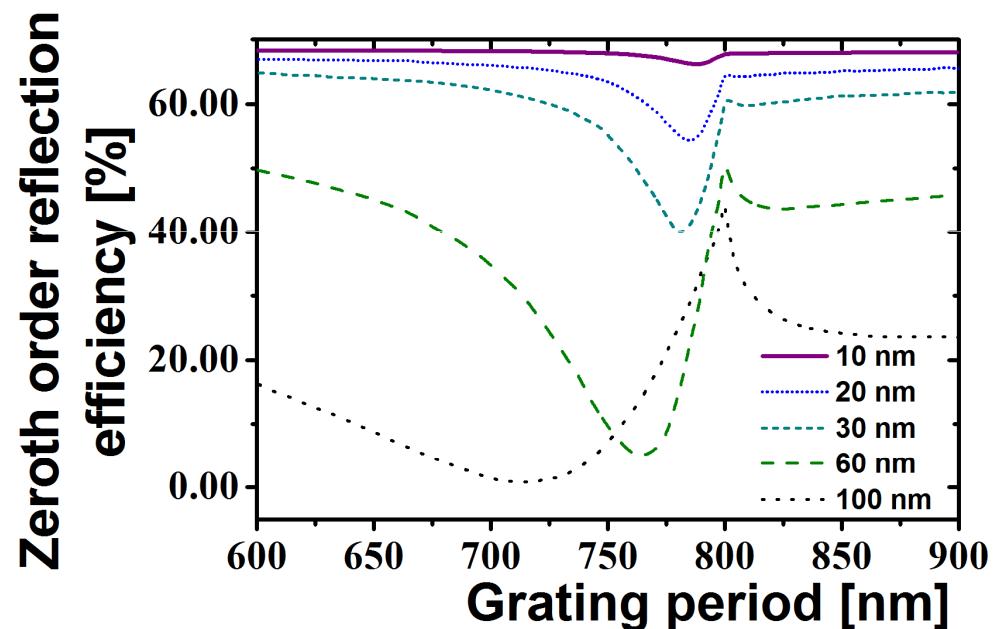
$$\tau_c / \tau_m \approx 20$$

$\nabla \gamma$ contributes to move the fluid
during the melt lifetime if $N > 20$

⇒ Resonance shifts
toward a smaller periodicity
for deeper gratings

⇒ May influence the final
morphology/periodicity

“Plasmonic feedback”: grating depth
can change the surface wave dispersion



Rigorous Coupled Wave Analysis (RCWA)
[Lyndin's code package at www.MCgrating.com]

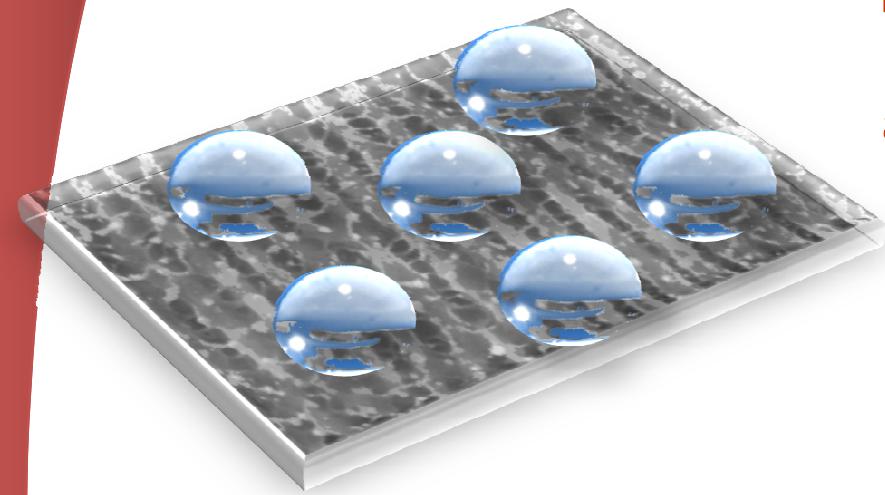
Conclusion: applications



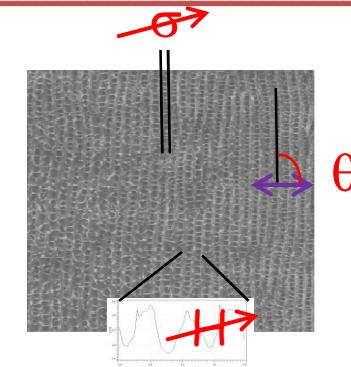
Controlling the topology

Tribology of
corrugated surfaces

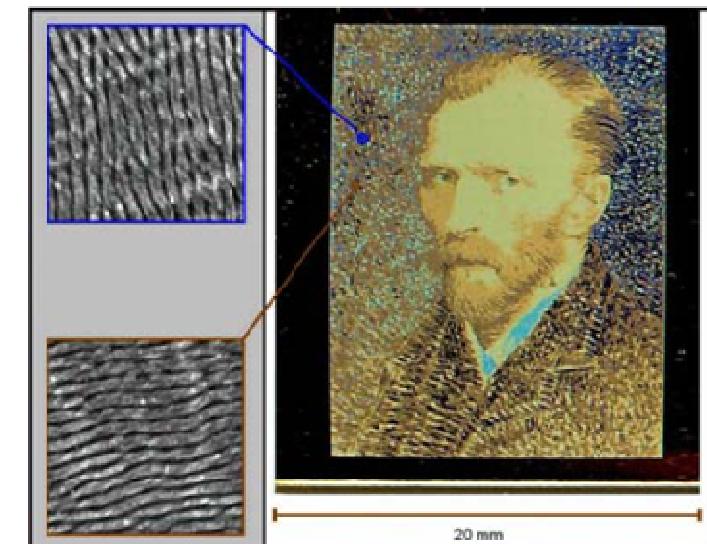
Super-hydrophobic
surfaces



Marking for
security
applications



Color coding and marking based on laser-induced nanostructures



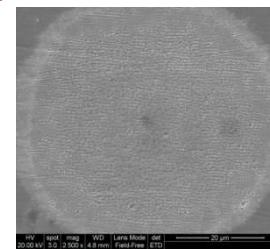
B. Dusser et al., Opt. Express 18, 2913 (2010)

Plasmonic & Hydro formation of fs LIPSS



*Cumulative Process
(N Laser shots)*

Positive feedback



100 fs

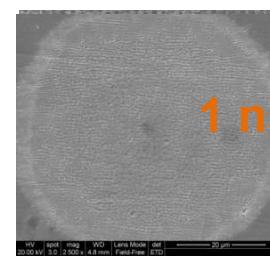
ps

100 ps

1 ns

time

Solidification



Conclusion

Ultrashort pulse

Absorption

CB

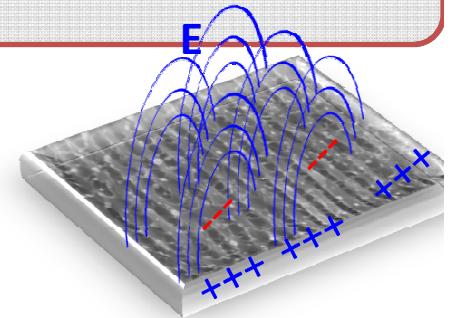
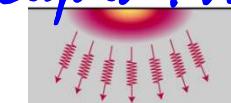
VB

e-ph coupling

CB

VB

Liquid Phase



Plasmonic experiment (periodicity)

(e-ph) coupling experiment (contrast)

Deepening of gratings
Thermocapillarity

Thanks for your attention !

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