First-Principles Calculations of Transient Optical Properties of Tungsten under Femtosecond Laser Irradiation

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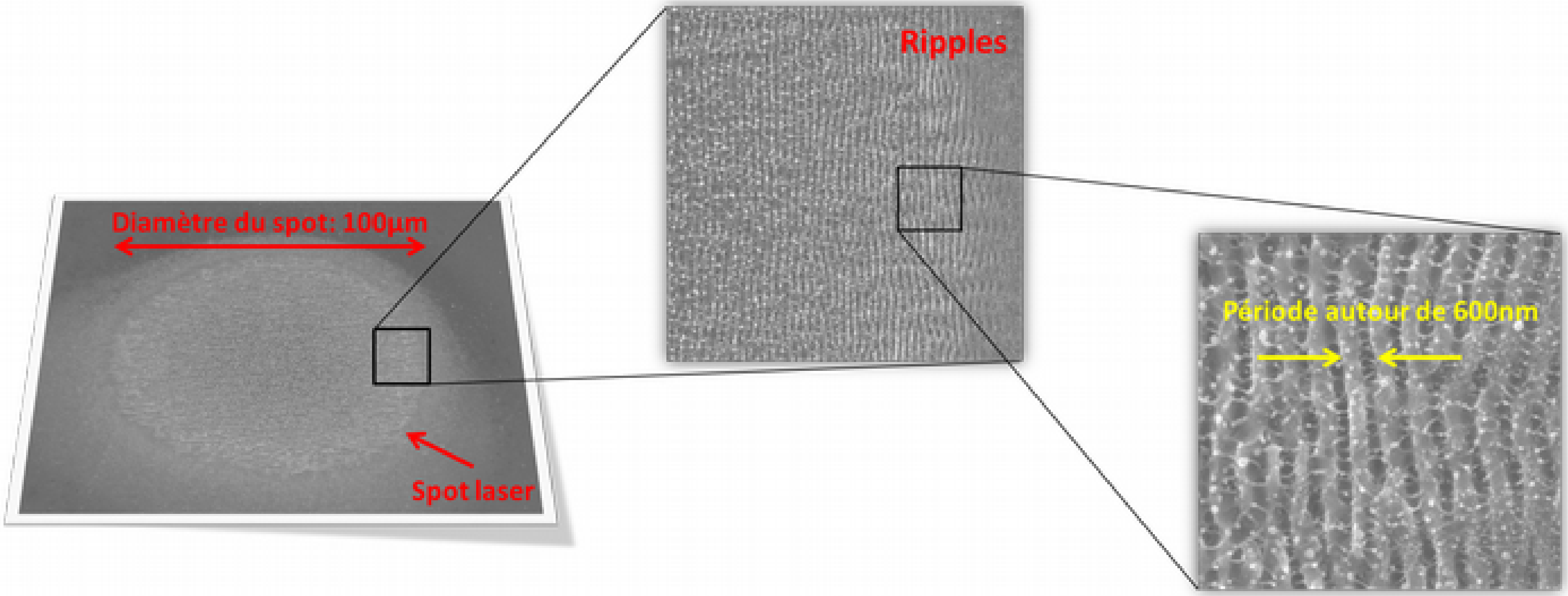
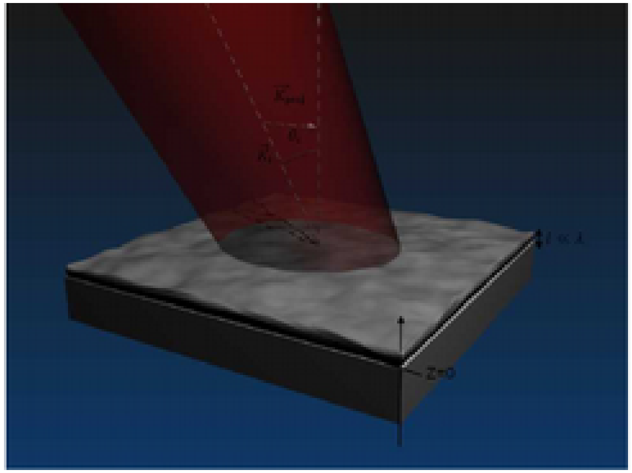
Ripples Formation

The irradiation of a sample to ultrashort laser impulsions lead to the formation of periodic structures called ripples or LIPSS (Laser Induced Periodic Surface Structures)



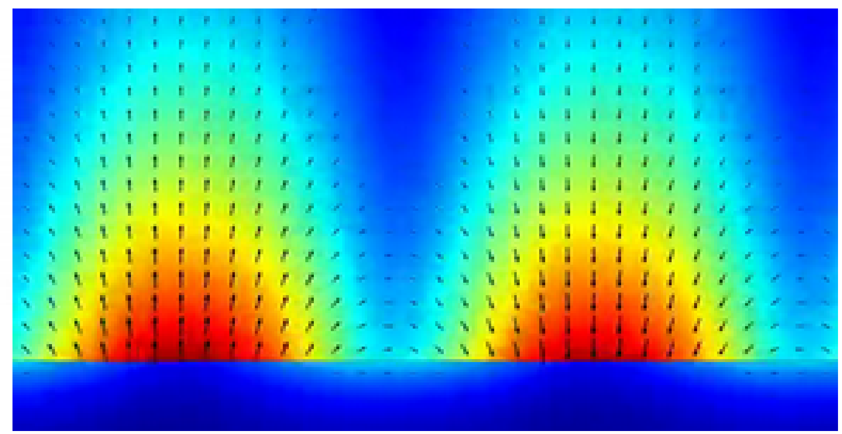
Interference model

Laser irradiation



Can excite Surface Plasmon
when interacting with the matter

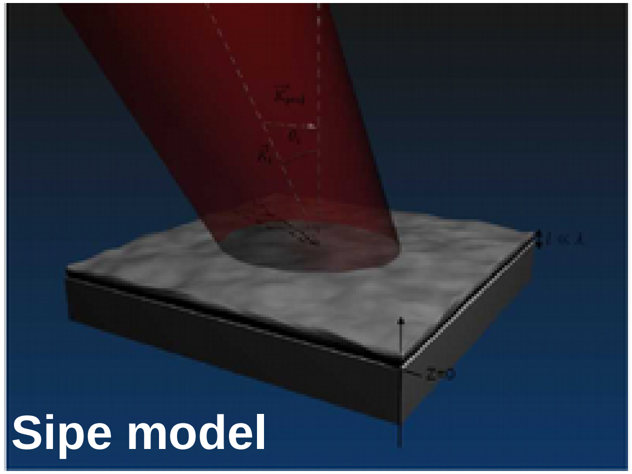
Surface Plasmon:
Oscillation of charges



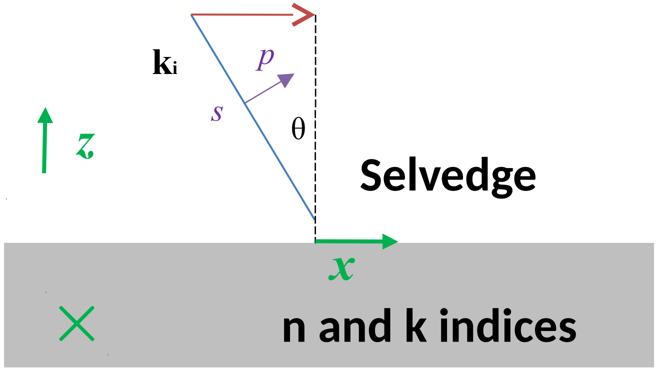
Given periodicity

Matter rearrangement according to Surface Plasmon periodicity

Electrodynamic model and predictions

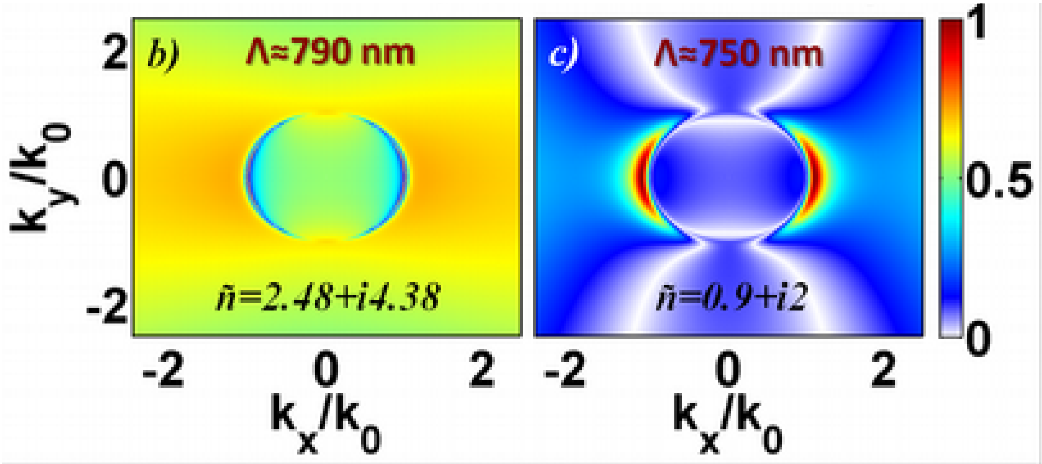


J. E. Sipe, Phys. Rev. B **27**, 1141 (1983).



Surface-scattered field interfere with the incident beam

From optical indices and the use of electrodynamic models, it is possible to deduce plasmonic excitation and thus ripple formation



Disagreement between SP and ripples periodicities

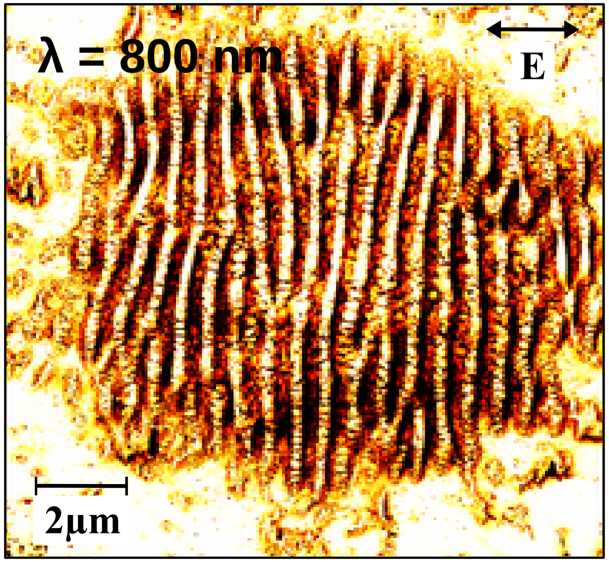
Change of optical properties during the irradiation process?

The tungsten, a non plasmonic metal having ripples

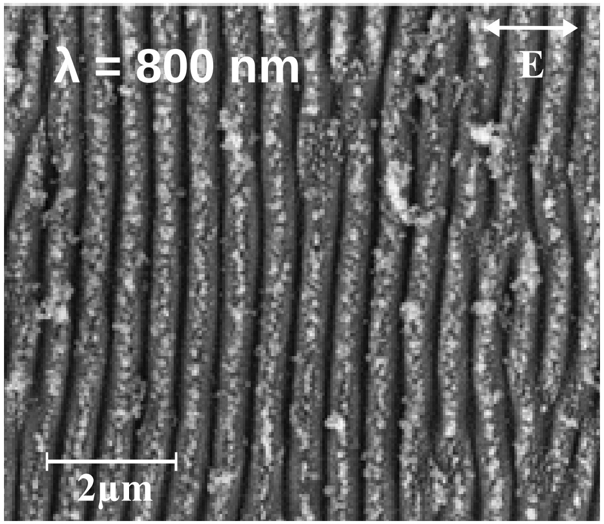
Standard optical indices at λ = 800 nm: n = 3.6 and k = 2.7 preclude the condition for plasmonic excitation $n^2 - k^2 < -1$

→ According to interference model, ripples should not appear.

Disagreement with experimental observations :



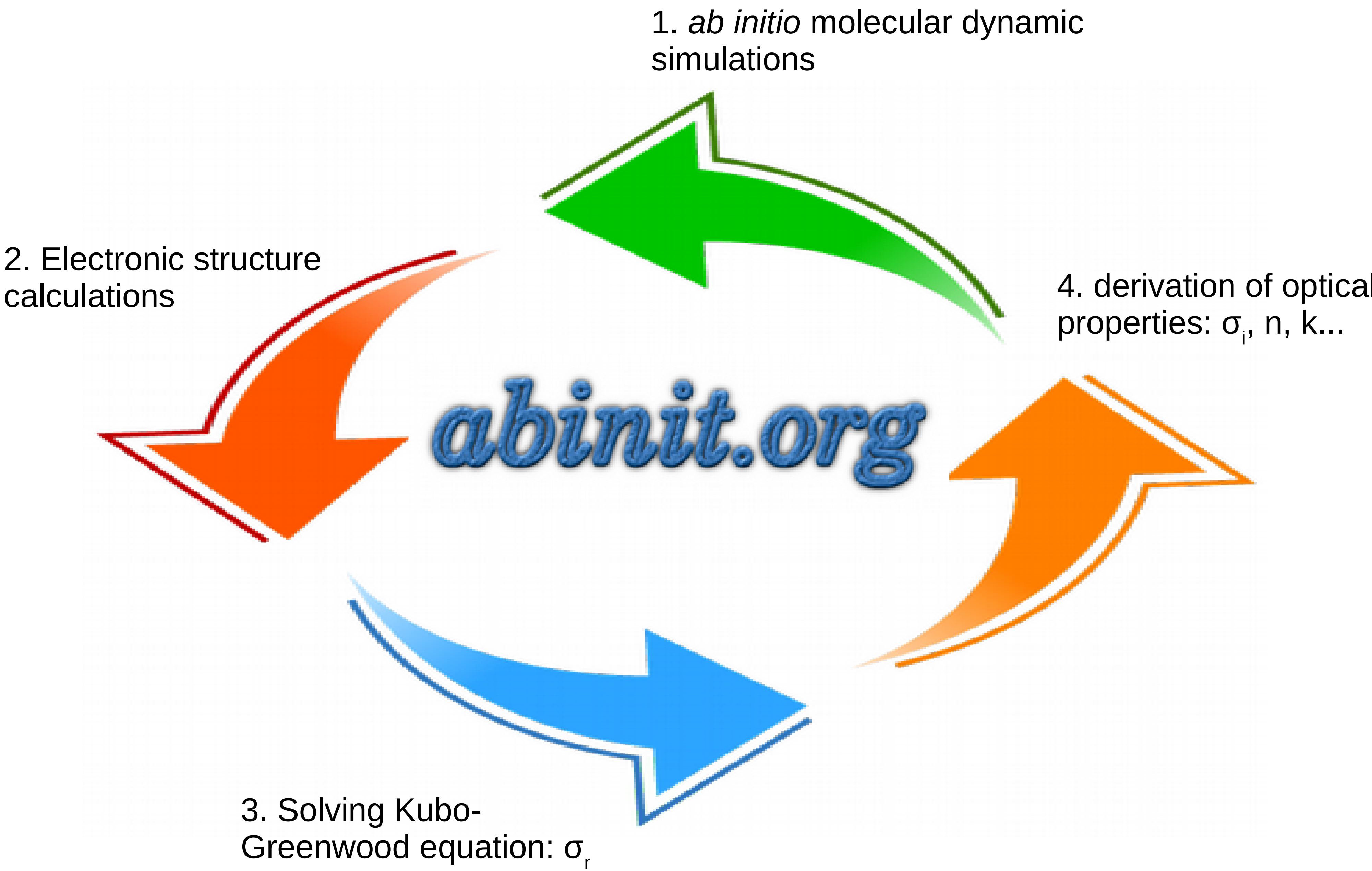
Reproduced in LaHC laboratory



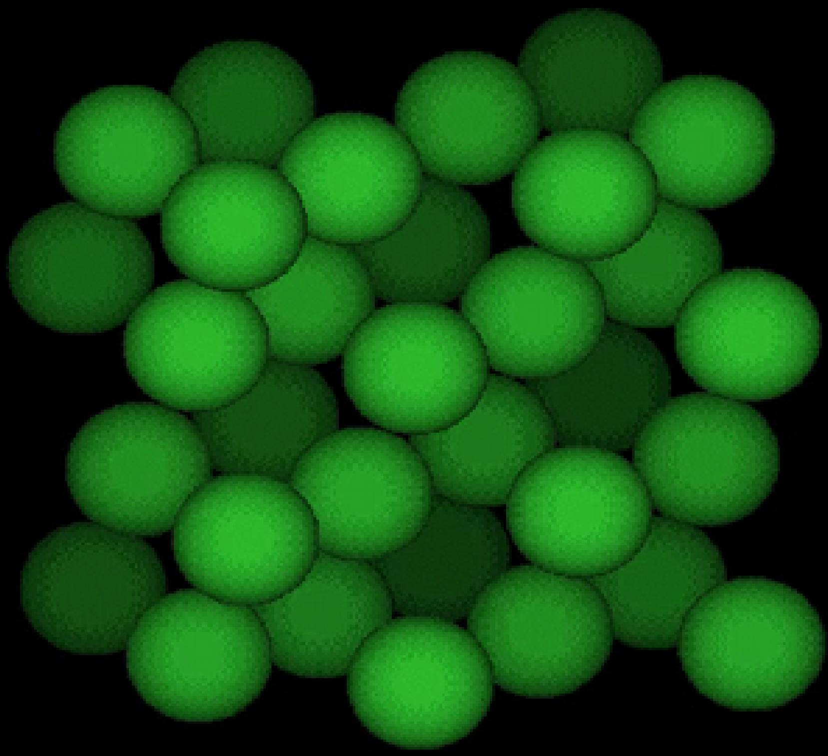
A. Y. Vorobyev and C. Guo, J. Appl. Phys. **104**, 063523 (2008)

Are optical properties changing enough to allow surface plasmon excitation?

Modelling of transient optical properties of tungsten



Performing *ab initio* molecular dynamic simulations



(Not a realistic MD!)

Tungsten case:

Crystal structure : BCC

Number of atoms: 54

Time step: ~2 fs

Total duration (2ps)

Temperature: ambient

Pressure: ambient

MD: isokinetic ensemble

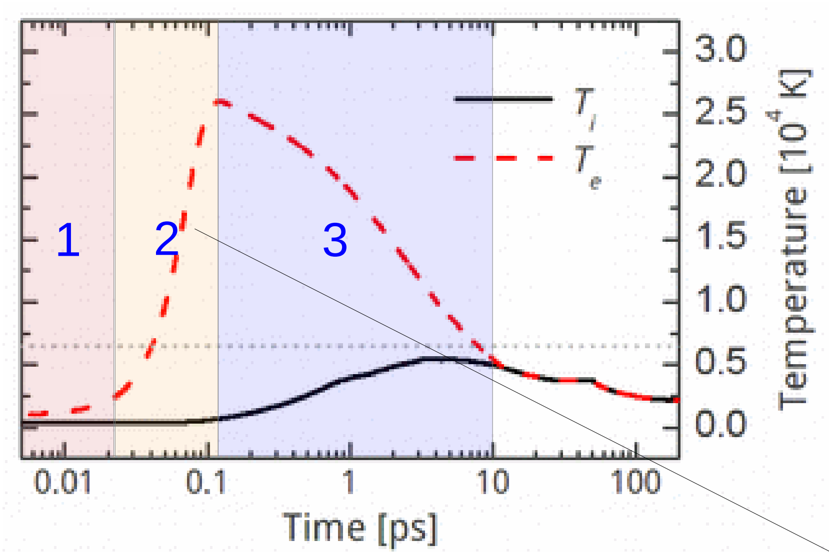
Visualization: Molden

Solving the equations of motion, forces are computed from the electronic structure calculation

Assuming a pseudo-equilibrium at the end of the MD

Ionic configurations considered as representative of ambient conditions

Electronic structure calculations, taking into account laser effect



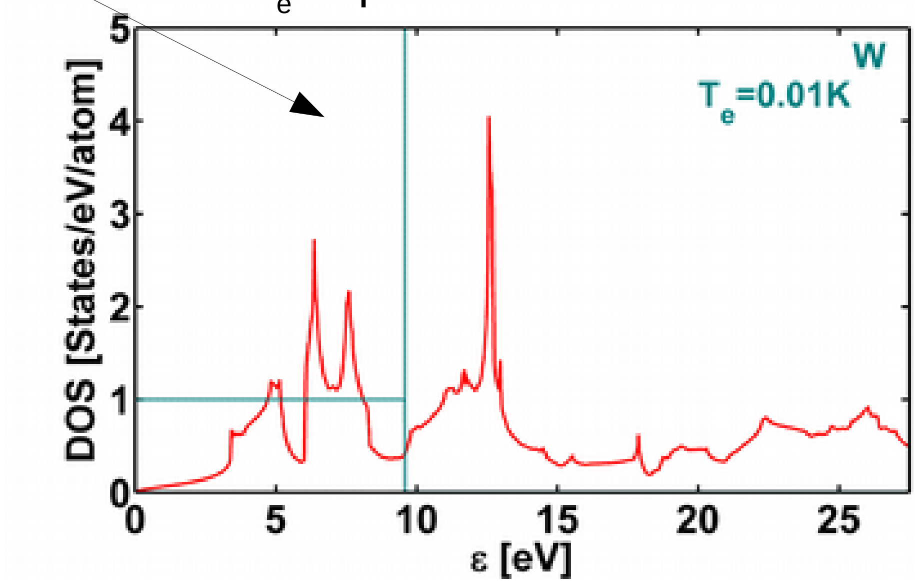
1: Unhomogeneous distribution of electrons (1-10 fs)

2: Thermalisation of electrons, from $e^- e^-$ collisions (10-100 fs)

3: Energy transfer from electron to the lattice

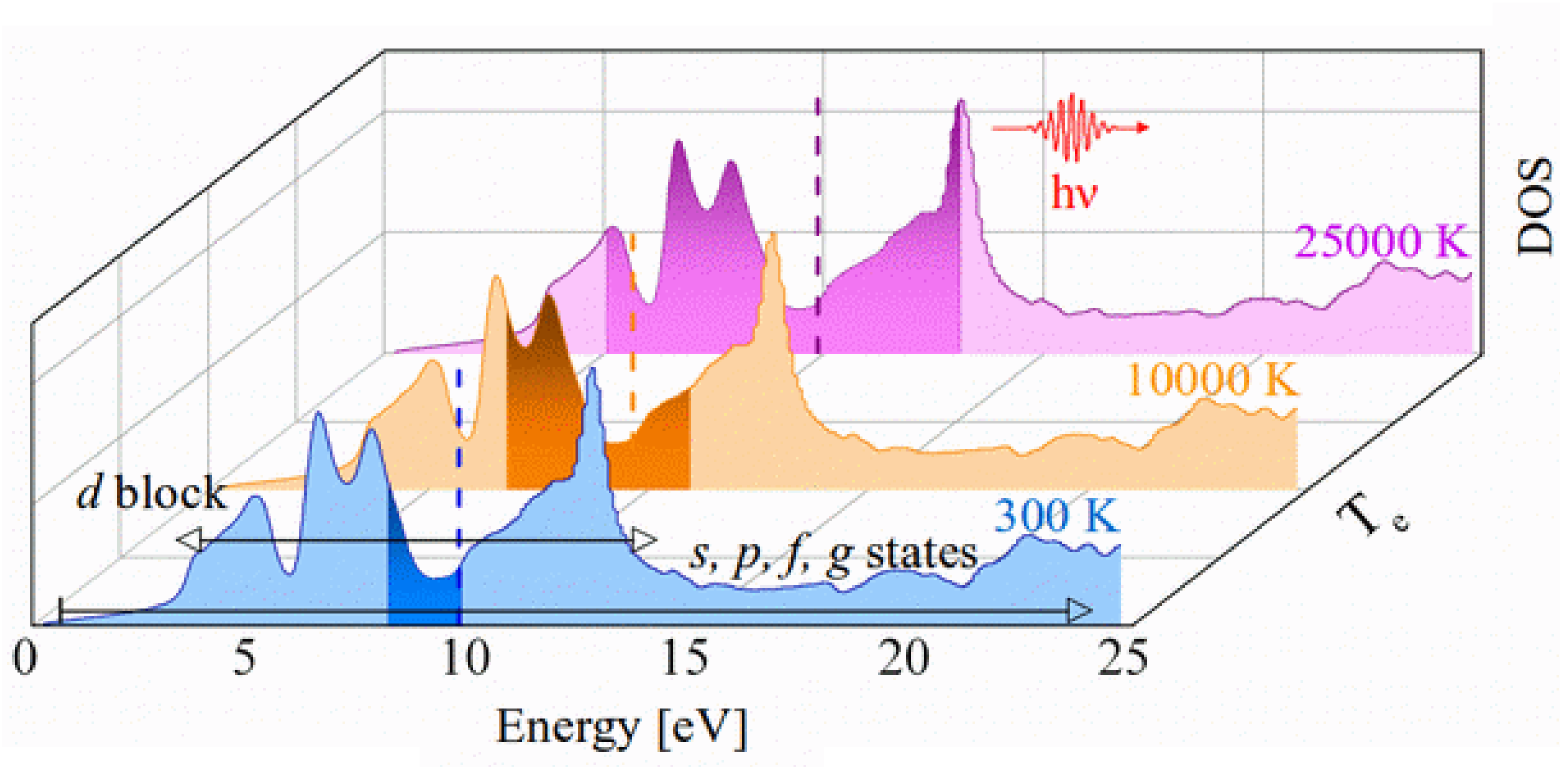
- ✓ *Density functional calculations*
- ✓ *Electronic temperature through:*
  - *Fermi-Dirac distribution of electrons*
  - *Minimization of Free energy: $F = E - T_e S_e$*
- ✓ *Laser effect taken into account by T_e dependent electronic structures*

T_e dependent DOS of W



E. Bevillon *et al.*, Phys. Rev. B **89**, 115117 (2014)

T_e dependent DOS of Tungsten: T_i = 300K, variable T_e



- ✓ *Stability of the electronic structure*
- ✓ *Electronic chemical potential locked into pseudo band gap*
- ✓ *Dark color: electronic states potentially involved in transition considering a photon energy of 1.55 eV (λ=800 nm) and Fermi broadening (3/2kbT$_e$).*

Time resolved optical properties according to the Kubo-Greenwood formalism and Kramers-Kroenig relation

$$\sigma_R(\omega)=\frac{2\pi e^2}{3\omega}\frac{1}{\Omega}\sum_k W_k \sum_{n,m}(f_n^k-f_m^k)\frac{1}{2\pi^2}|\langle\psi_n^k|\hat{\upsilon}|\psi_m^k\rangle|^2\delta(\epsilon_m^k-\epsilon_n^k-\hbar\omega)$$

$$\sigma_I(\omega)=\frac{-2}{\pi}\wp\int\frac{\sigma_{(\omega')}\omega}{\omega'^2-\omega^2}d\omega'$$

σ_R being the real part of the optical conductivity

ω being the pulsation considered

Ω being the volume of the cell

W_k being the *k*-point weight

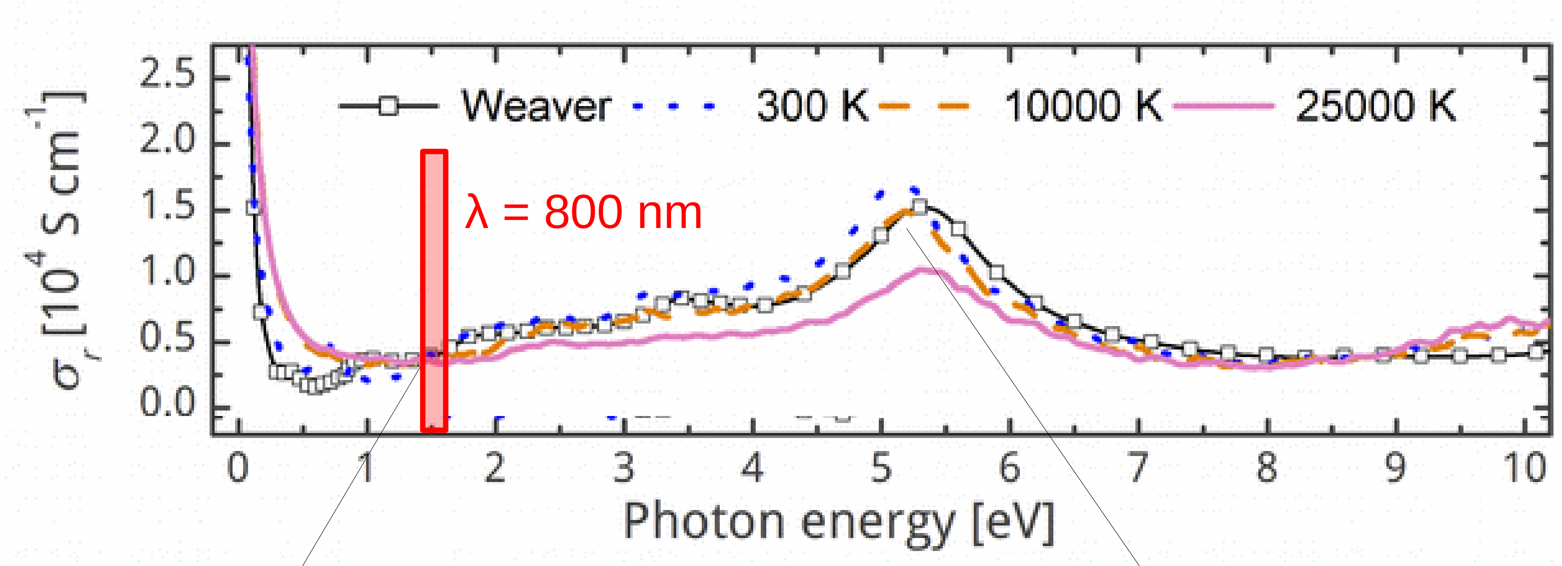
f_i^k being the occupation of the eigenstate i according to the Fermi-Dirac distribution

ψ_i^k being the eigenstate i at *k*-point k (wavefunction or band)

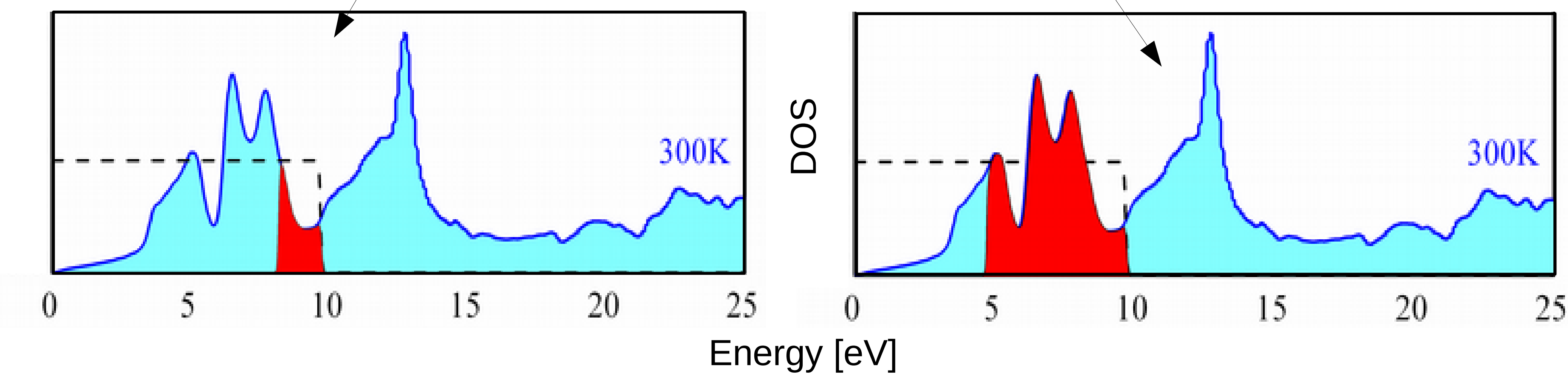
ϵ_i^k being the eigenvalue i at *k*-point k (energy of the wavefunction at this point)

$\wp$ being the principal value of the integral (Cauchy, improper integrals)

T_e dependent optical conductivities of tungsten



Fermi factor: electronic states potentially involved in electronic transition



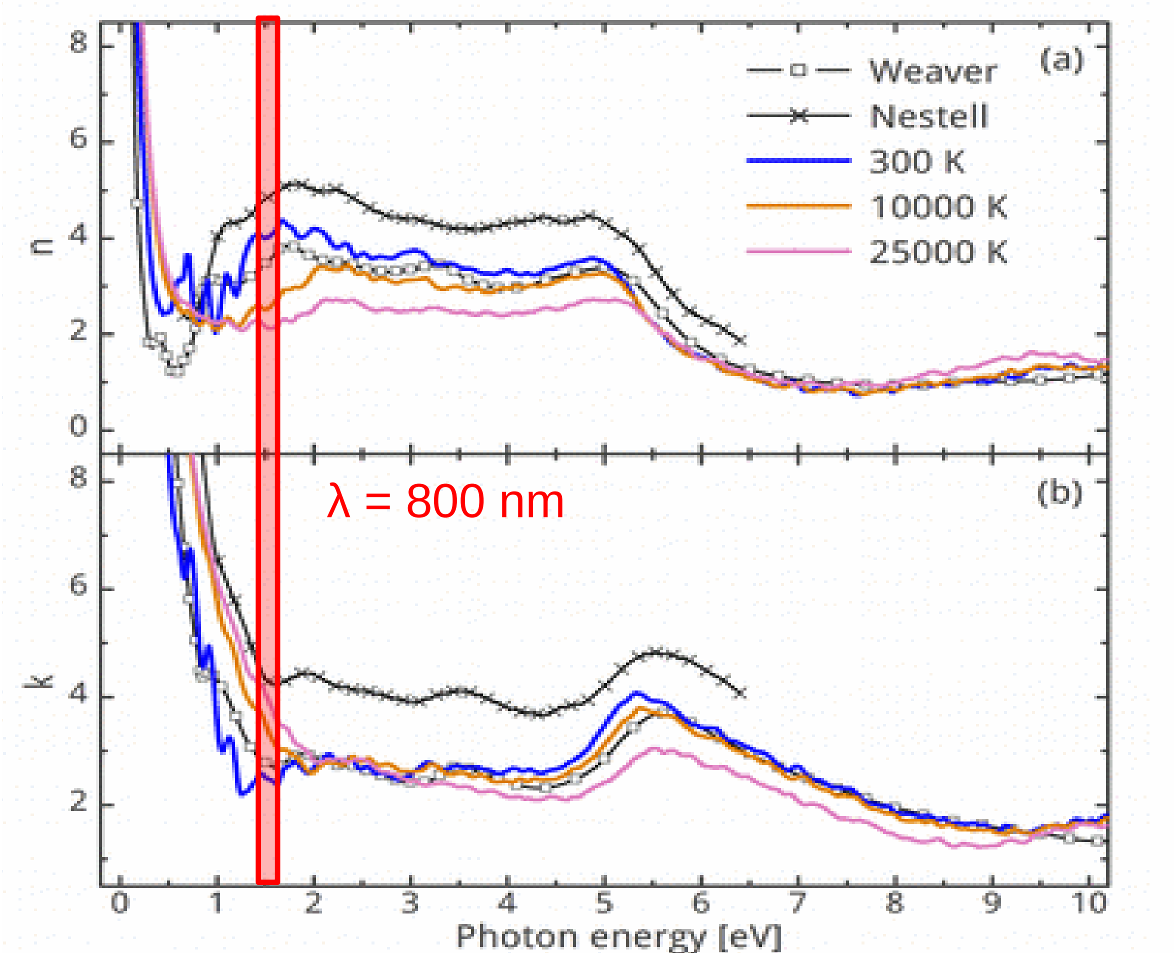
- ✓ *Increase of intraband and attenuation of interband signals from “dilution” of electronic transitions*

T_e dependent optical indices of tungsten

$$\begin{cases} \epsilon_r = 1 - \dfrac{\sigma_i}{\omega \epsilon_0} \\ \epsilon_i = \dfrac{\sigma_r}{\omega \epsilon_0} \end{cases}$$

$$\begin{cases} n = \left[\left(\epsilon_r^2 + \epsilon_i^2 \right)^{1/2} + \epsilon_r \right]^{1/2} \\ k = \left[\dfrac{\left(\epsilon_r^2 + \epsilon_i^2 \right)^{1/2} - \epsilon_r}{2} \right]^{1/2} \end{cases}$$

→ Complex mix of σ_r and σ_i



- ✓ *Changes dependent on the photon energy*
- ✓ *At λ = 800 nm:*
  - *n significantly decreases*
  - *k significantly increases*

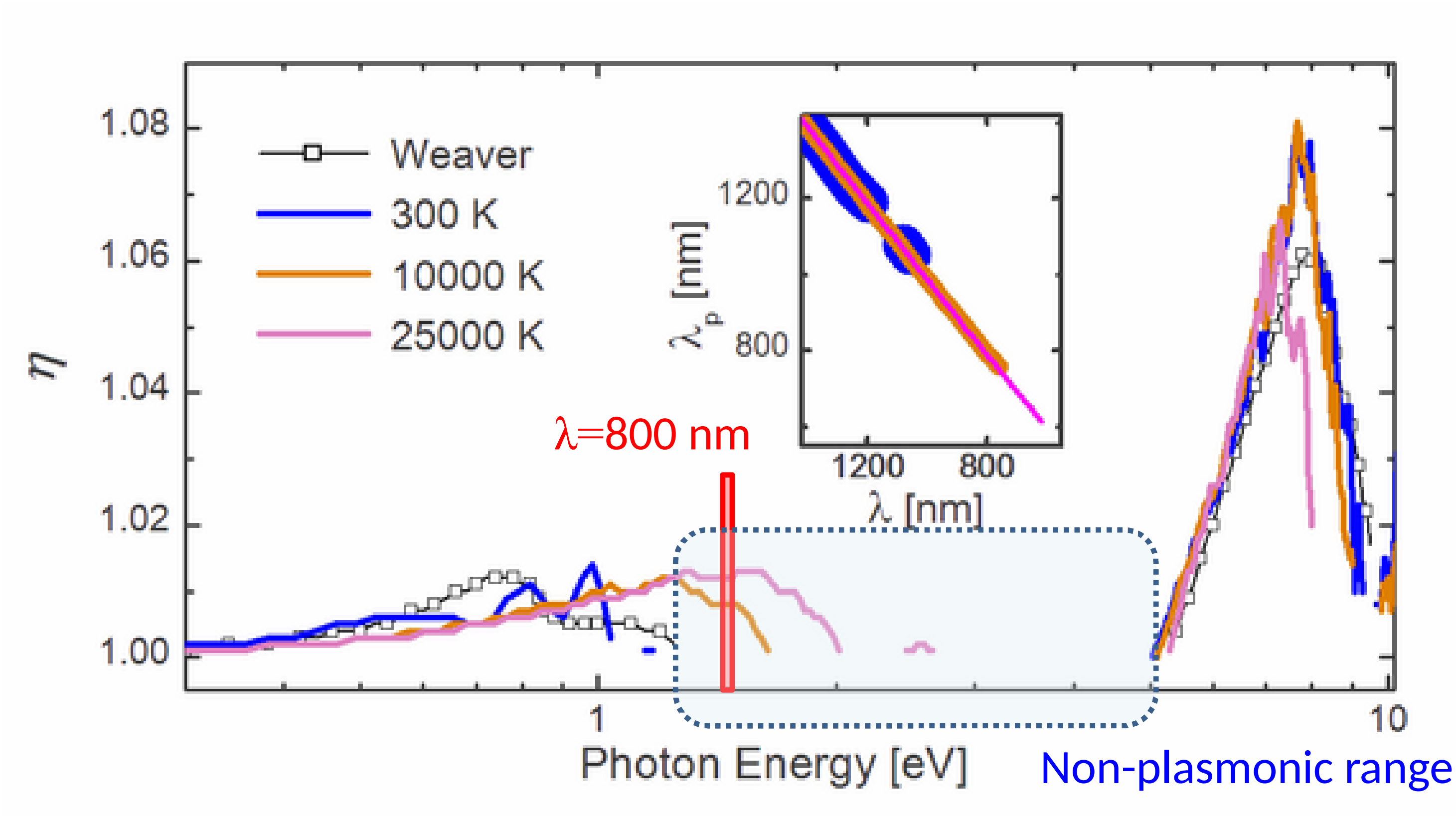
Surface plasmon existence domain

$$\lambda_{SP} = \lambda / \eta$$

with $\eta = \Re\{[\tilde{n}^2/(\tilde{n}^2+1)]^{1/2}\}$

Condition to excite SP wave:

$$n^2 - k^2 < -1$$



T_e increase:

- ✓ *Clear extension of the existence domain*
- ✓ *Plasmonic switching at 800 nm for high T_e*
- ✓ $\eta = \lambda / \lambda_{SP}$ *does not evolve significantly (1 or 2%)*

Conclusion

- ✓ *Optical properties are changing during the irradiation process*
- ✓ *Changes attributed to decrease of interband signal induced by Fermi-broadening dilution of d-bands transition*
- ✓ *Significant impact on optical indices*
- ✓ *Modification of the existence domain of the surface plasmon*
- ✓ *Not a significant modification of SP wavelengh, the disagreement between theory and experience may still rely on geometrical factors*
- ✓ *Surface effects or phase instabilities may also affect optical properties*

Thank you for attention