Understanding Ripples Formation: Nonequilibrium Properties of Stainless Steel and Tungsten Metal

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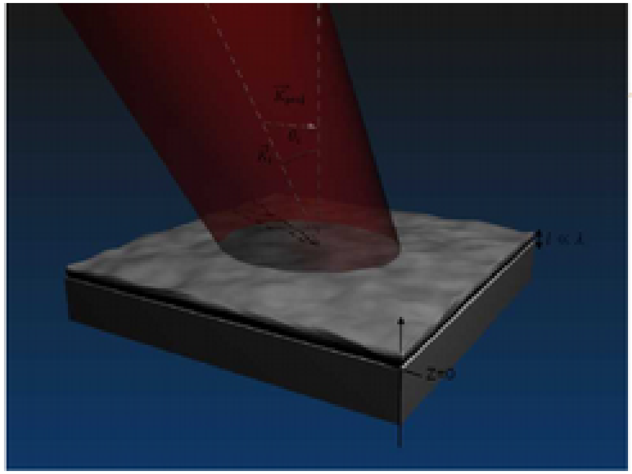
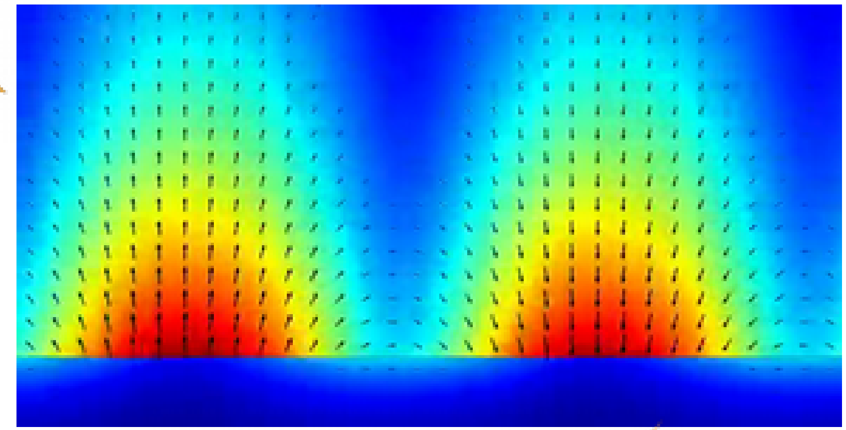
In the Framework of the Interference Model

Ultrashort laser irradiation

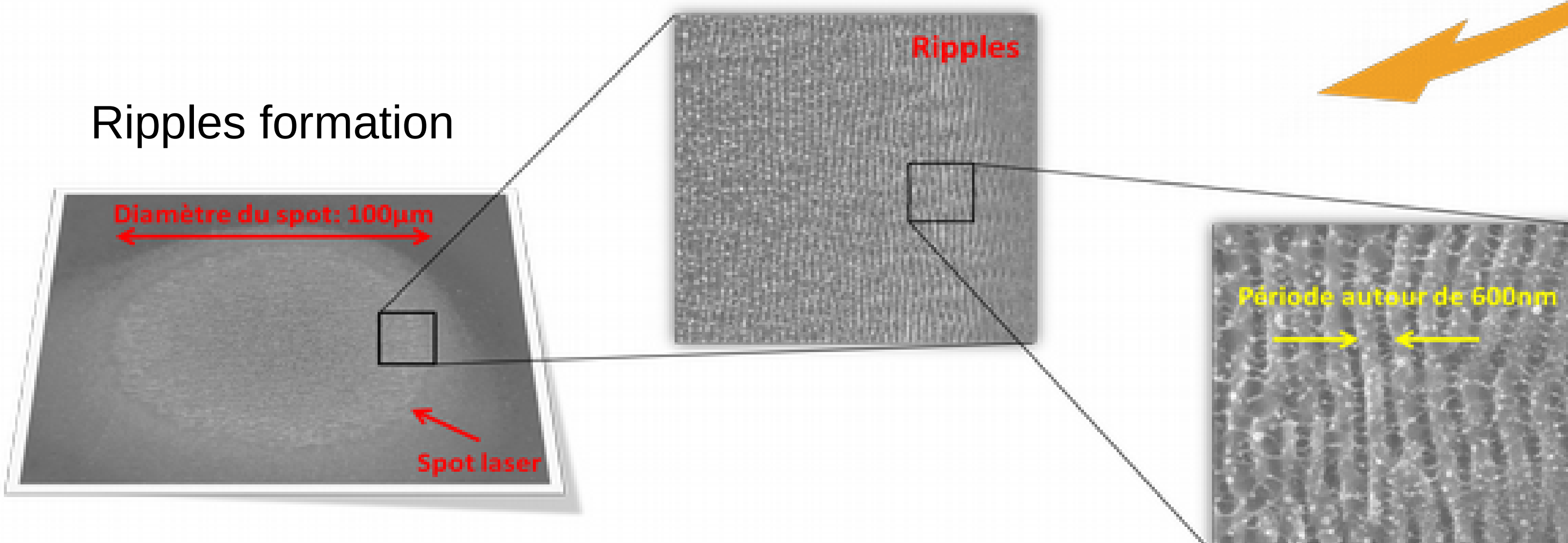


$$n^2 - k^2 < -1$$

Surface Plasmon: Oscillation of charges

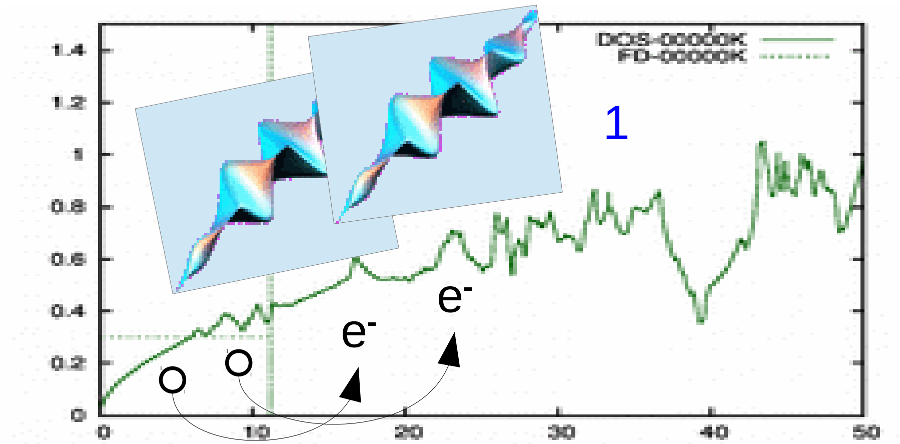


Ripples formation



Reorganization of the matter with plasmon periodicity

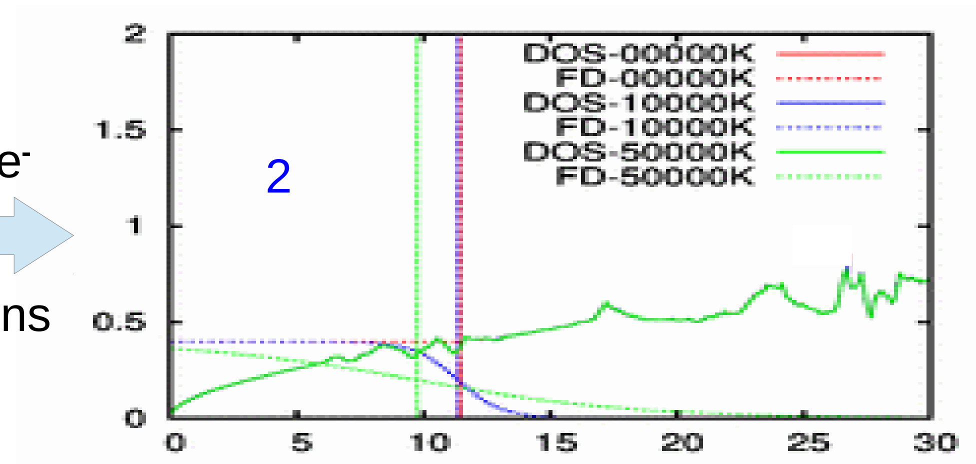
Electronic Structures & Laser Irradiation



Unhomogeneous excitation of electrons

$e^- <-> e^-$

Collisions

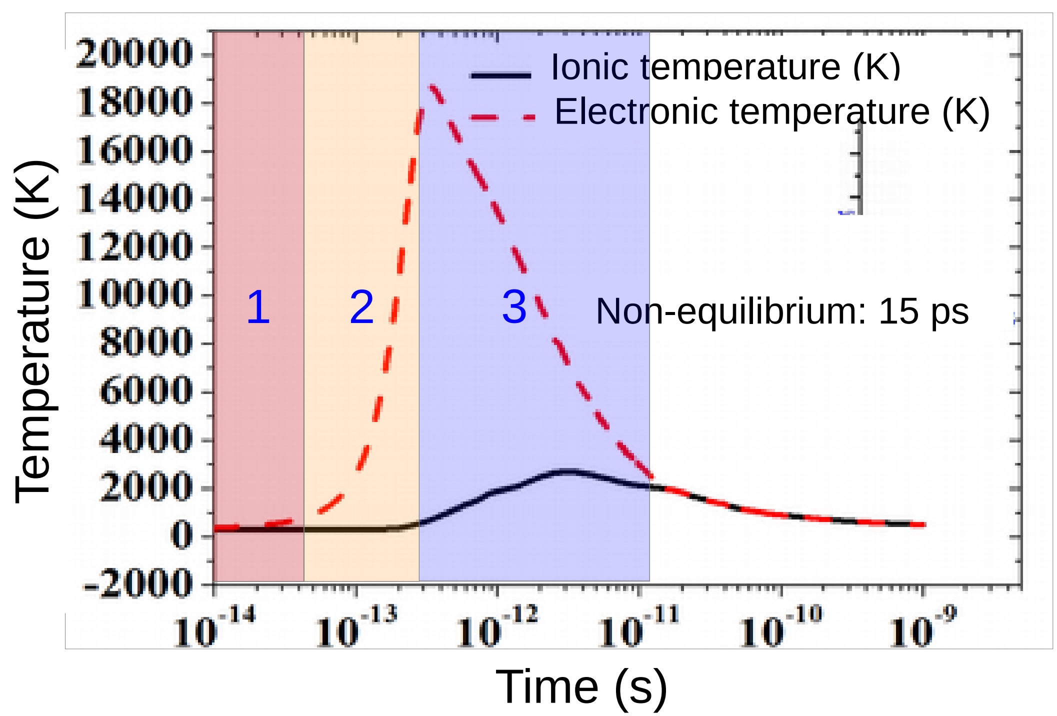


Thermalized state, described by electronic temperature (T_e)

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

2: Thermalization of electrons, homogeneous distribution from electronic collisions (10-100 fs)

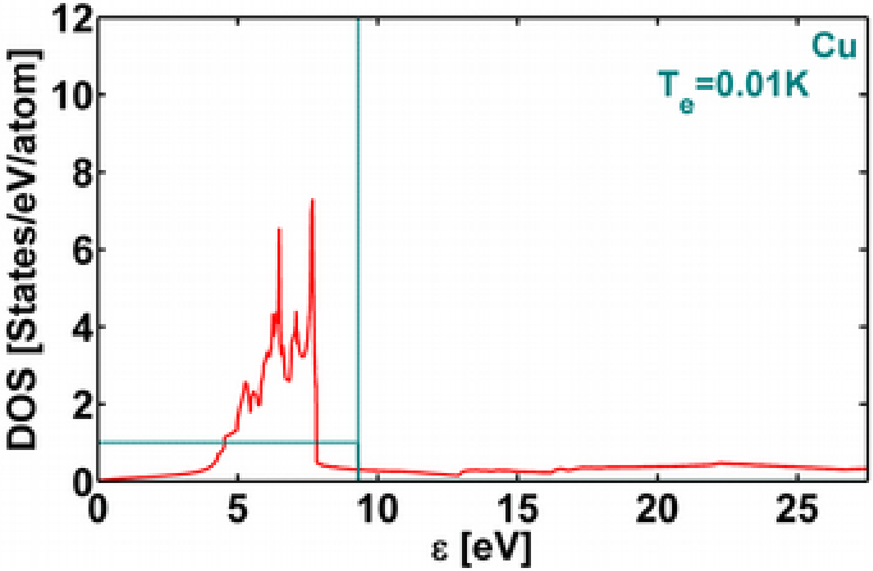
3: Electrons-ions energy exchanges, from election-ion coupling (ps)



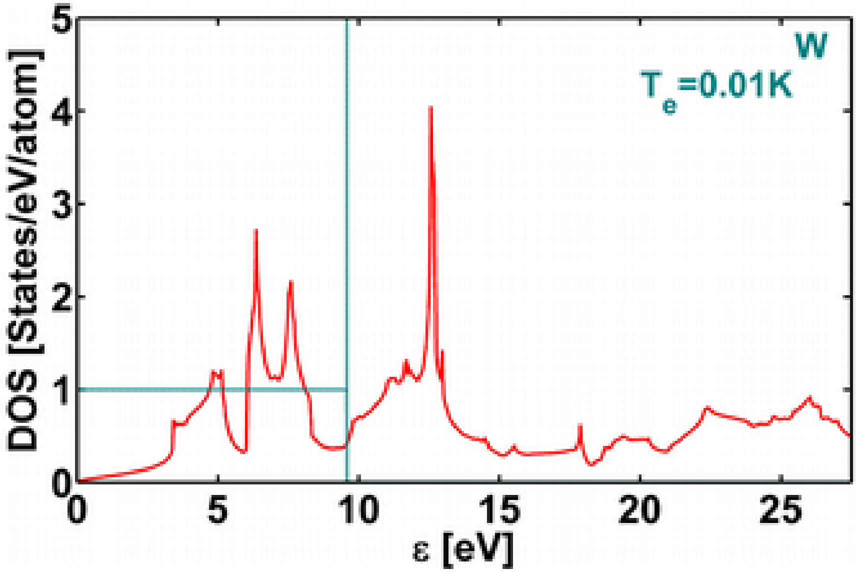
Finite Temperature Density Functional Theory Calculations

- *FT – DFT calculations to account for nonequilibrium conditions*
- *Fermi Dirac distribution function for electrons*
- *Self-consistent computation of T_e dependent electronic structures*

T_e dependent DOS of Cu

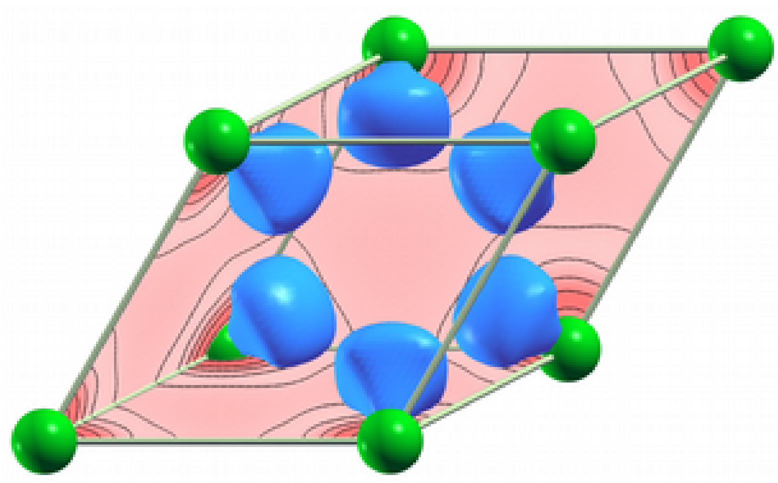


T_e dependent DOS of W

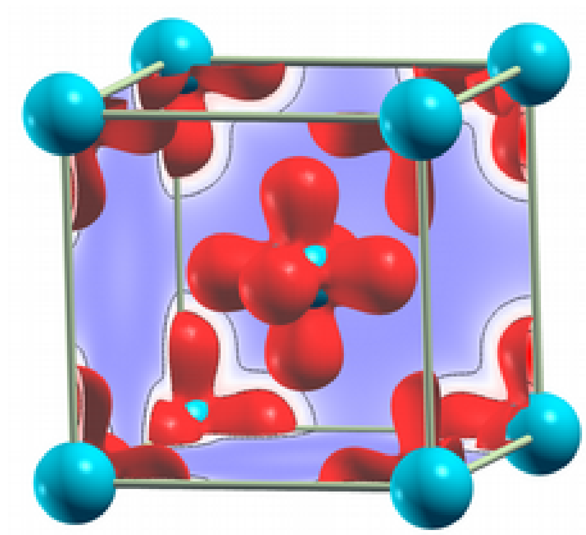


E. Bevillon *et al.*, Phys. Rev. B **89**, 115117 (2014)

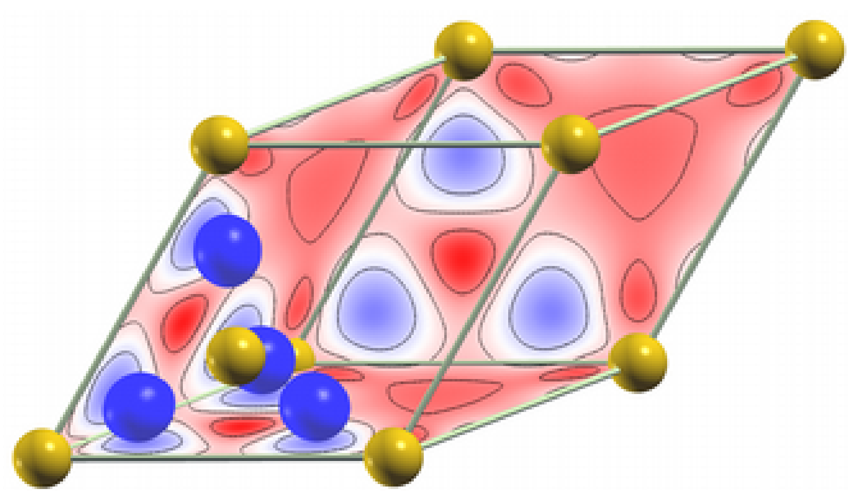
Electronic Density Differences Between Hot and Cold Densities,
(mapping and isosurfaces)



Ni, primitive FCC cell



Cr, conventional BCC cell



Si, primitive diamond cell

Xcrysden images:
Blue: loss of electronic density
Red: gain of electronic density

→ Impact on material properties:
thermodynamics, optics, athermal forces..

Tungsten Nonequilibrium Optical Properties

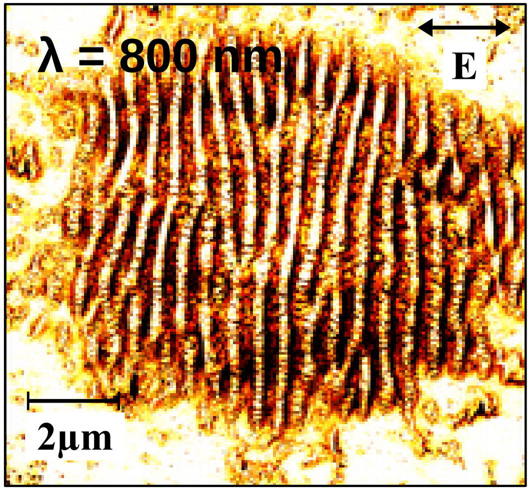
Tungsten, a Non-Plasmonic Metal Showing Ripples Formation

At λ = 800 nm: n = 3.6 ; k = 2.7
preclude the condition for plasmonic excitation $n^2 - k^2 < -1$

→ According to interference model, ripples should not appear.

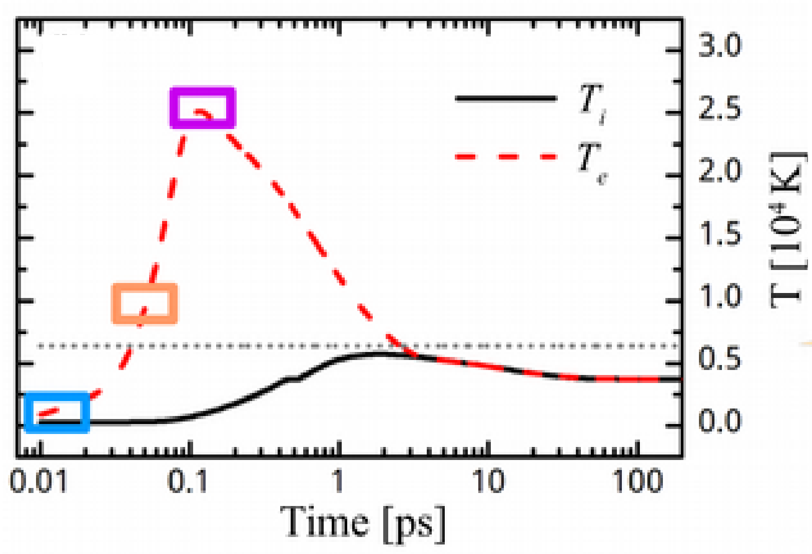
Disagreement with experimental observations :

Are optical properties changing during the irradiation process?



→ MD-FT-DFT coupled to KG formalism to determine temperature dependent optical properties.

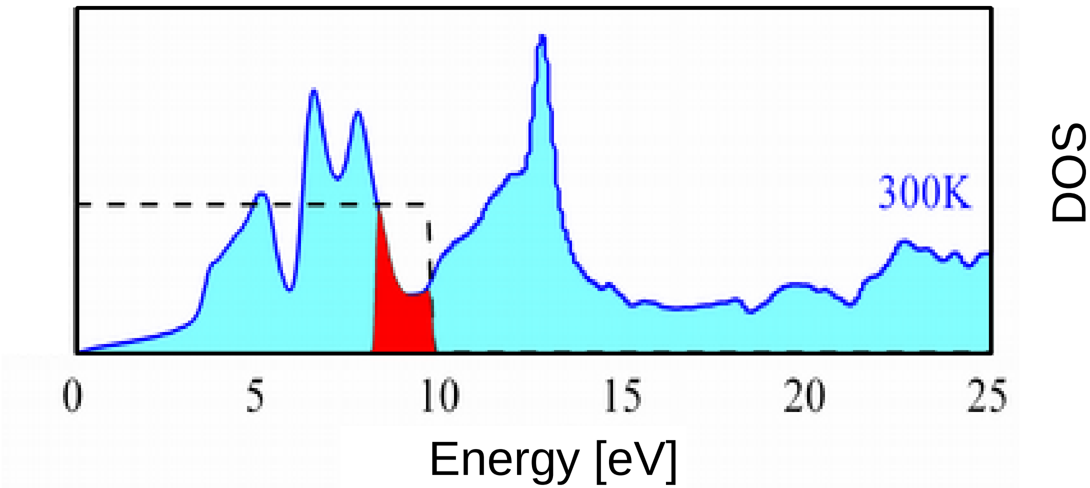
Temperature Evolutions of Irradiated Tungsten and its Effect on Potential Electronic Transitions



- *Two temperature model of W for an ultrashort laser irradiation close to damage threshold.*

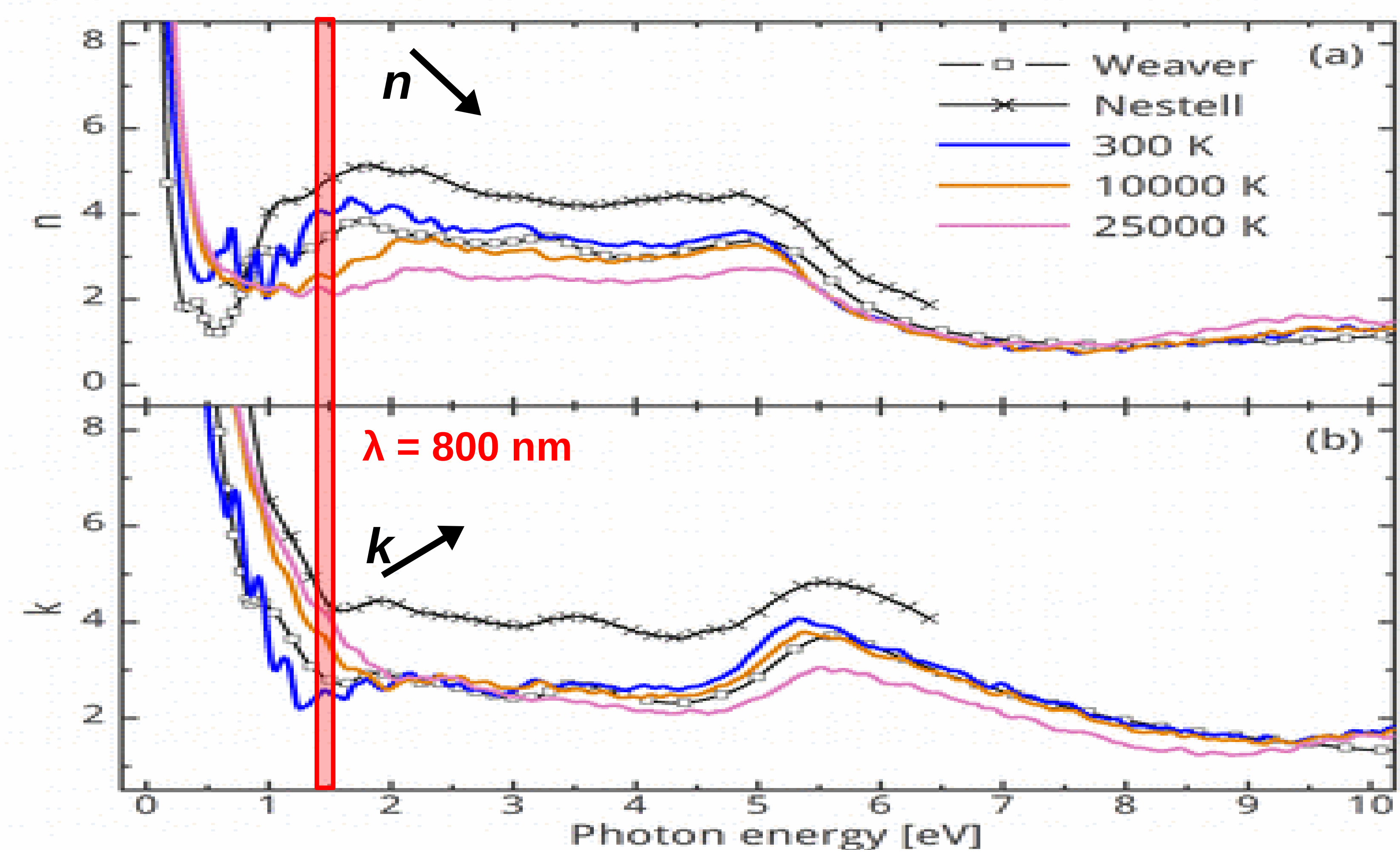
→ *T_i ~ ambient ; Variable T_e*

Potential electronic transitions at λ = 800 nm



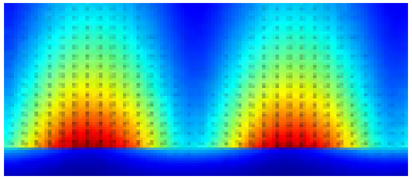
- *W electronic structure at T_i = 300K*
- *Increase and dilution of the electronic transition space phase with T_e increases*
- *Impact on tungsten optical properties*

T_e Dependent Optical Indices of Tungsten



- *Increase of intraband signal, decrease of interband signal*
- *At λ = 800 nm: n significantly decreases ; k significantly increases*

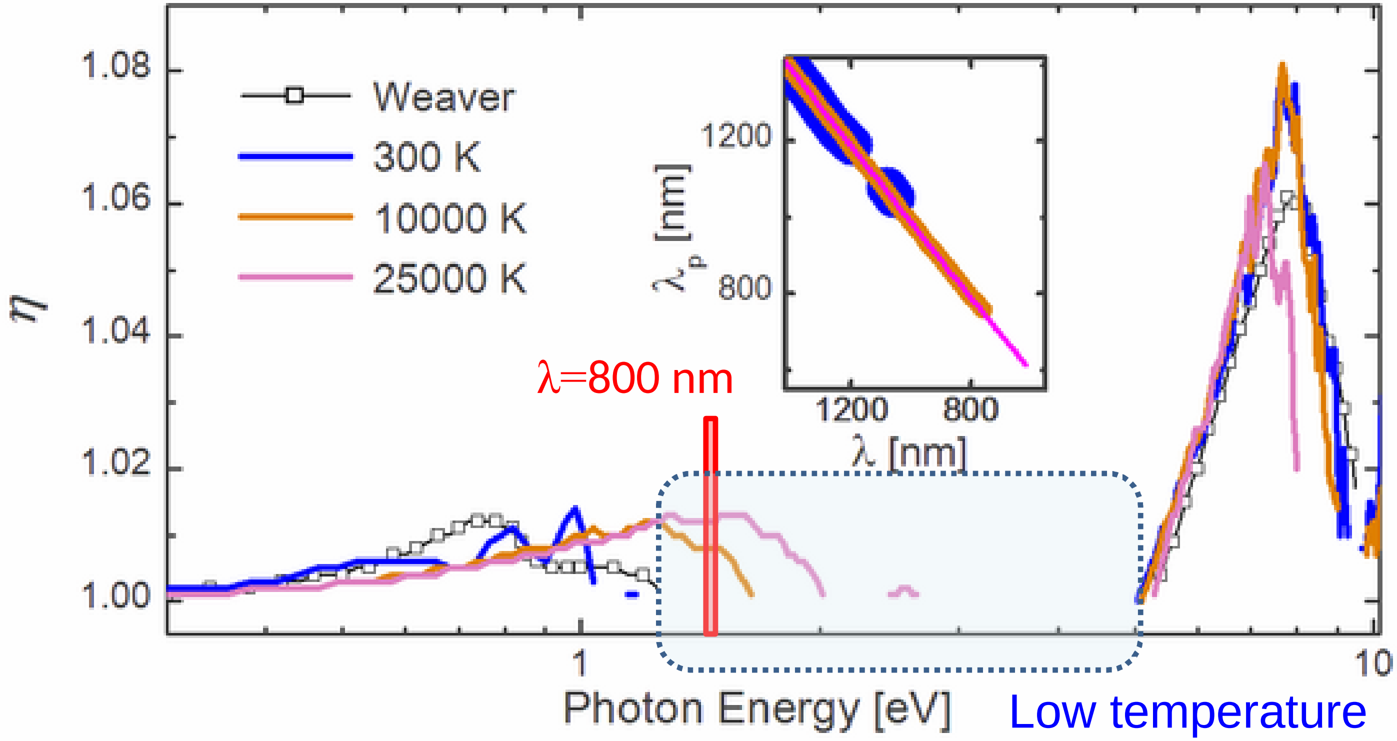
Surface Plasmon Existence Domain



$$\lambda_{SP} = \lambda / \eta \qquad \text{with} \qquad \eta = \Re\{[\tilde{n}^2/(\tilde{n}^2+1)]^{1/2}\}$$

Condition to excite SP wave:

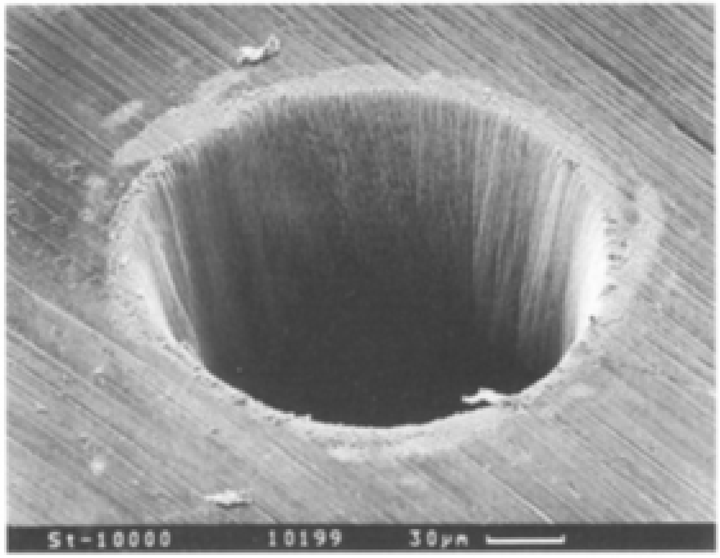
$$n^2 - k^2 < -1$$



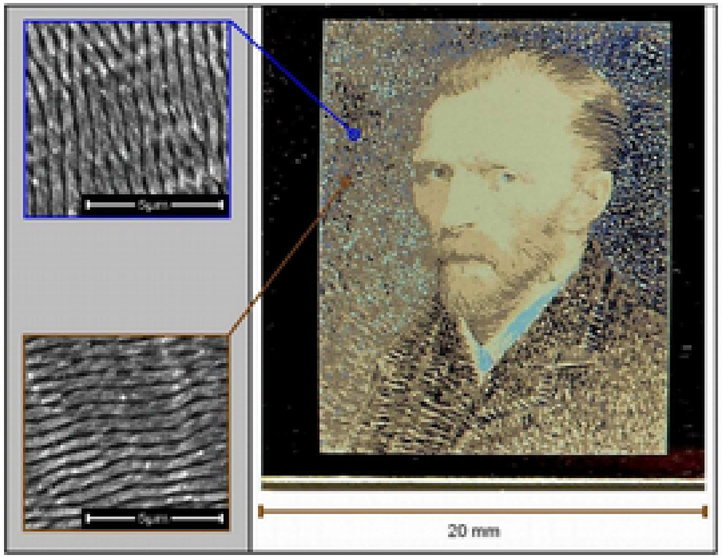
- *Extension of the plasmon existence domain*
- *Plasmonic switch at 800 nm at high T_e*
- *Agreement between Interference model and experiments*

316L Stainless Steel Nonequilibrium Properties

Stainless Steel Surface Structuring Under Ultrashort Laser Irradiation



B. Chichkov, et al. Appl. Phys. A **63**, 109–115 (1996).

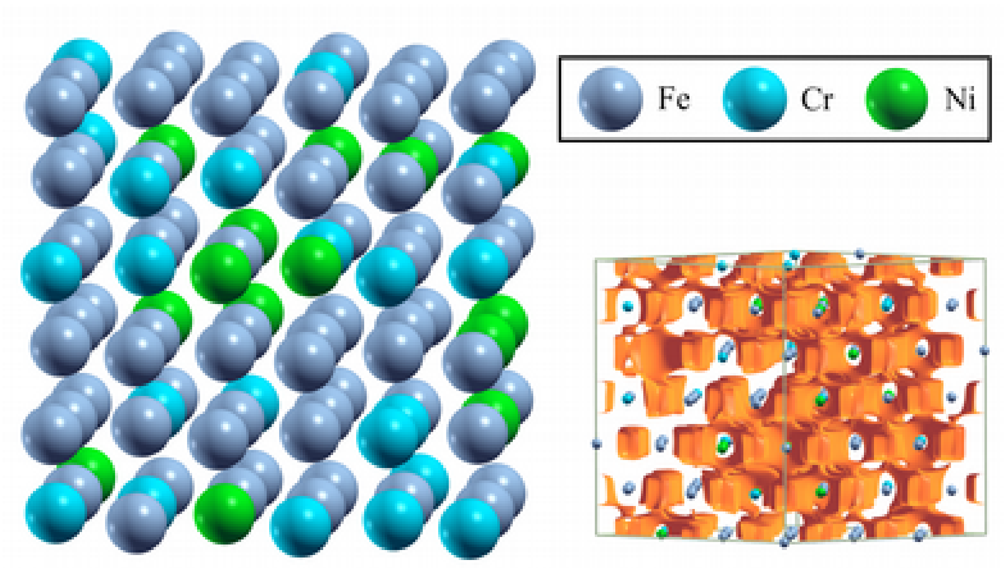


B. Dusser, et al. Opt. Express 18, 2913-2924 (2010).

- *Leads to numerous applications in optical functionalization, tribology or wettability*
- *Knowledge of nonequilibrium behavior is required*
- *Lack of theoretical data from atomistic approaches*

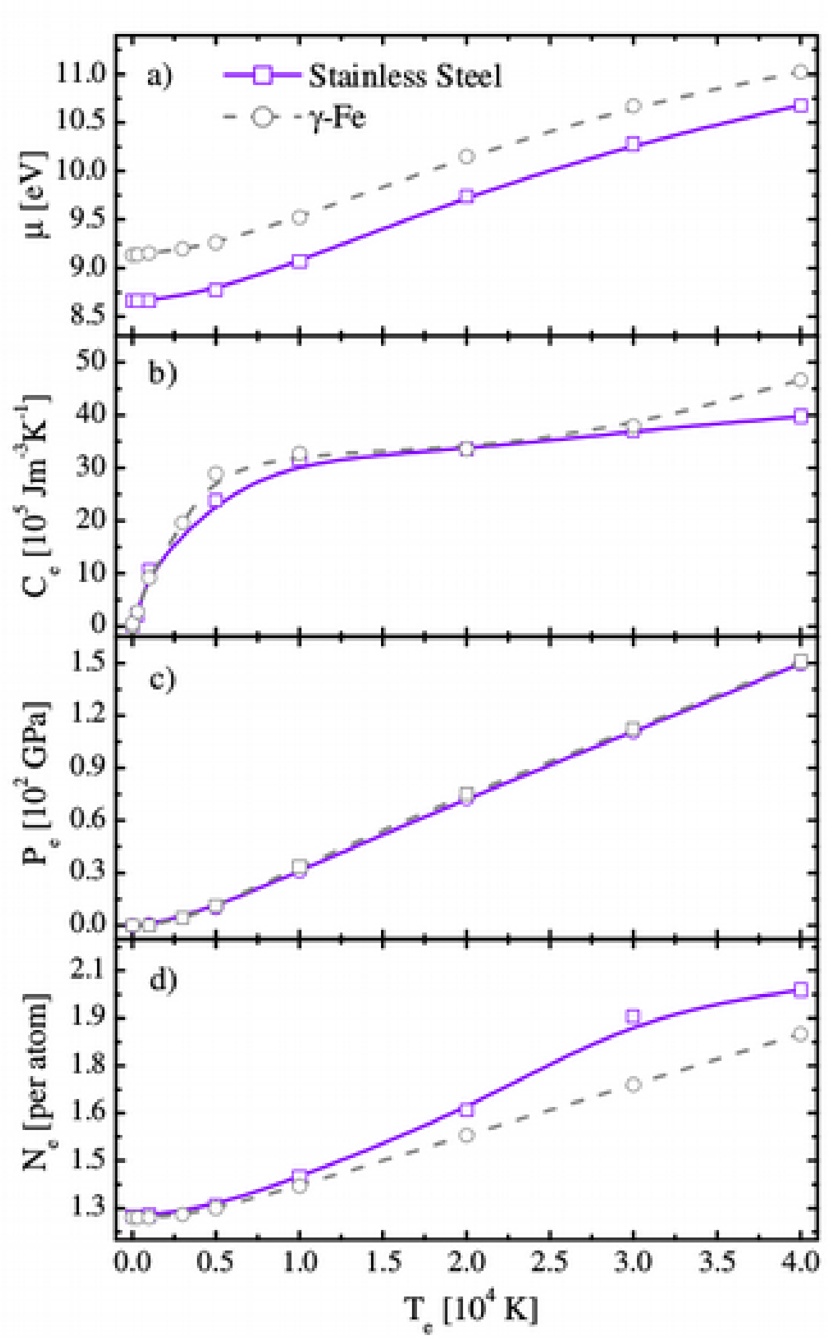
316L Stainless Steel Characteristics

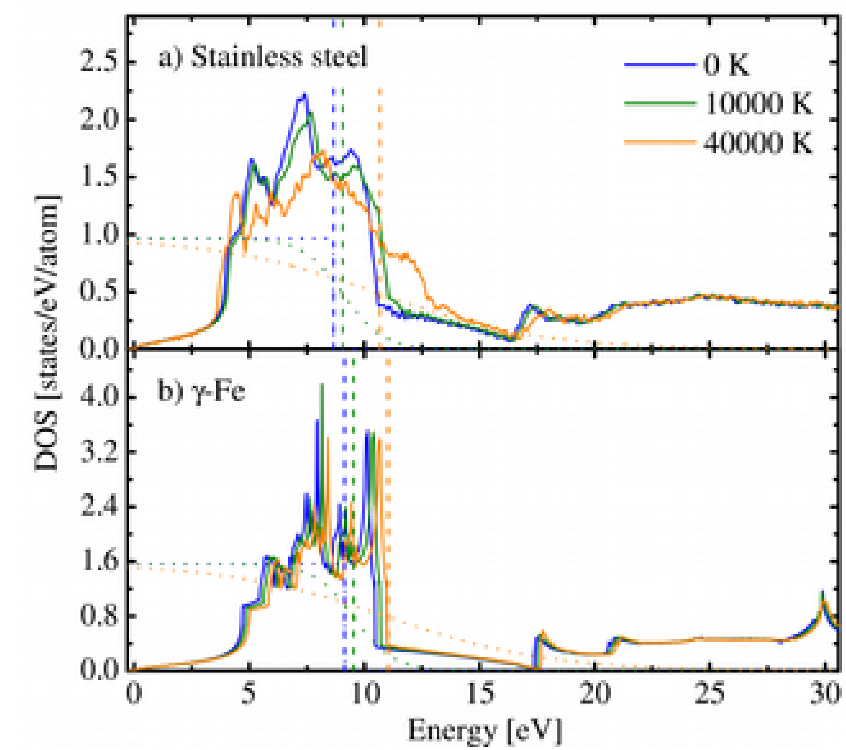
- *Austenitic steel, with FCC arrangement of metal elements in solid solution*
- *Mass composition: Fe (66%), Cr (17%), Ni (12%), Mo (2%), Mn (2%), Si (1%), P (0.04%), S (0.03%) and C (0.02%)*
- *Approximated to main constituents, Fe (68%), Cr (18%) and Ni (14%)*
- *Special Quasi Random distribution of atoms*



E. Bévillon et al. J.Phys. Chem. C (2015)
doi: 10.1021/acs.jpcc.5b02085.

Nonequilibrium Thermodynamic Properties of 316L Stainless Steel

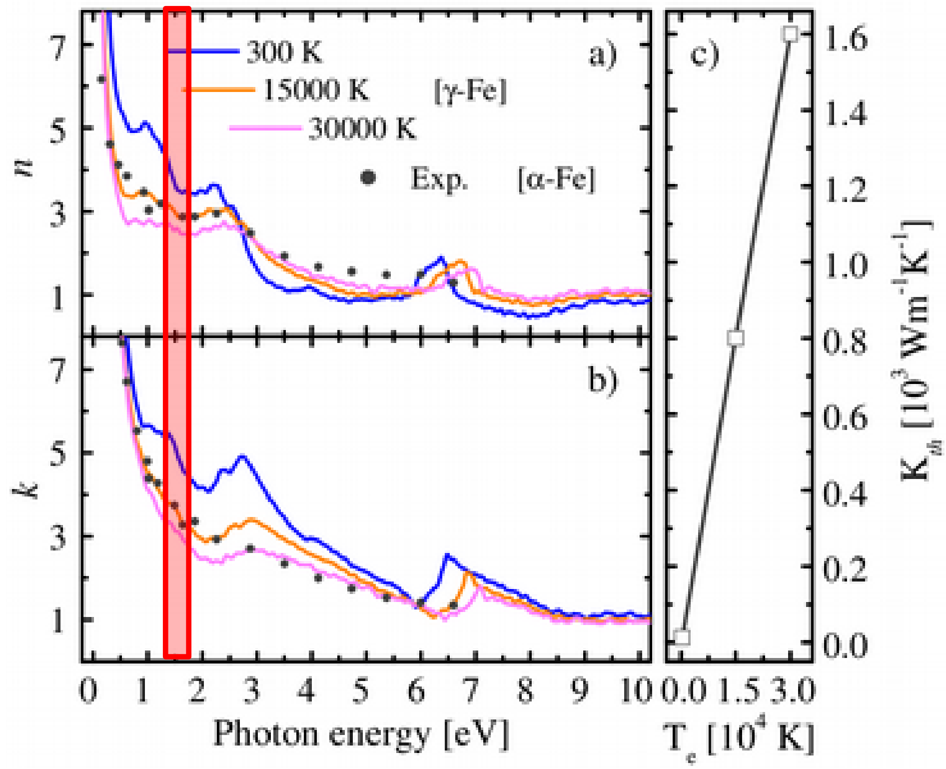




- T_e dependent DOS, d-block enlarging and shifting toward high temperature
- T_e dependent μ_e, C_e, P_e and N_e determined
- *Strong similarities with γ-Fe.*

Nonequilibrium Transport Properties of γ-Fe (Standing for Stainless Steel)

λ = 800 nm *Optical Properties*

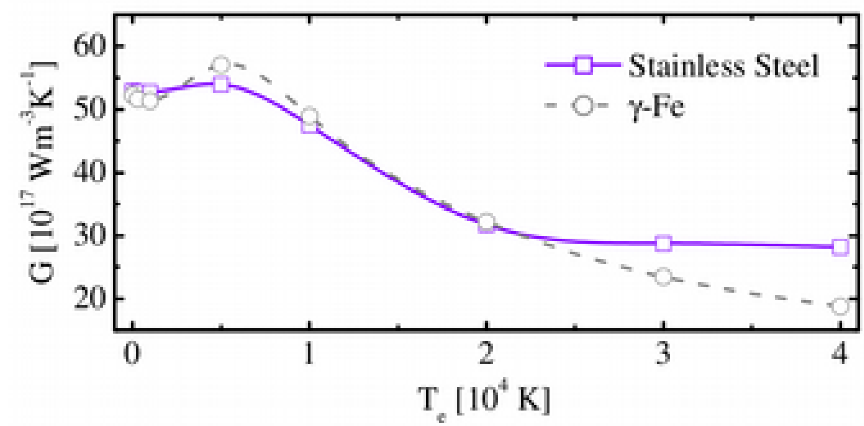


KG-MD + FT-DFT calculations

(T_i = 300K ; variable T_e)

- *n and k decrease*
- *K_{th} strongly increases*

Effective Electron-Phonon Coupling



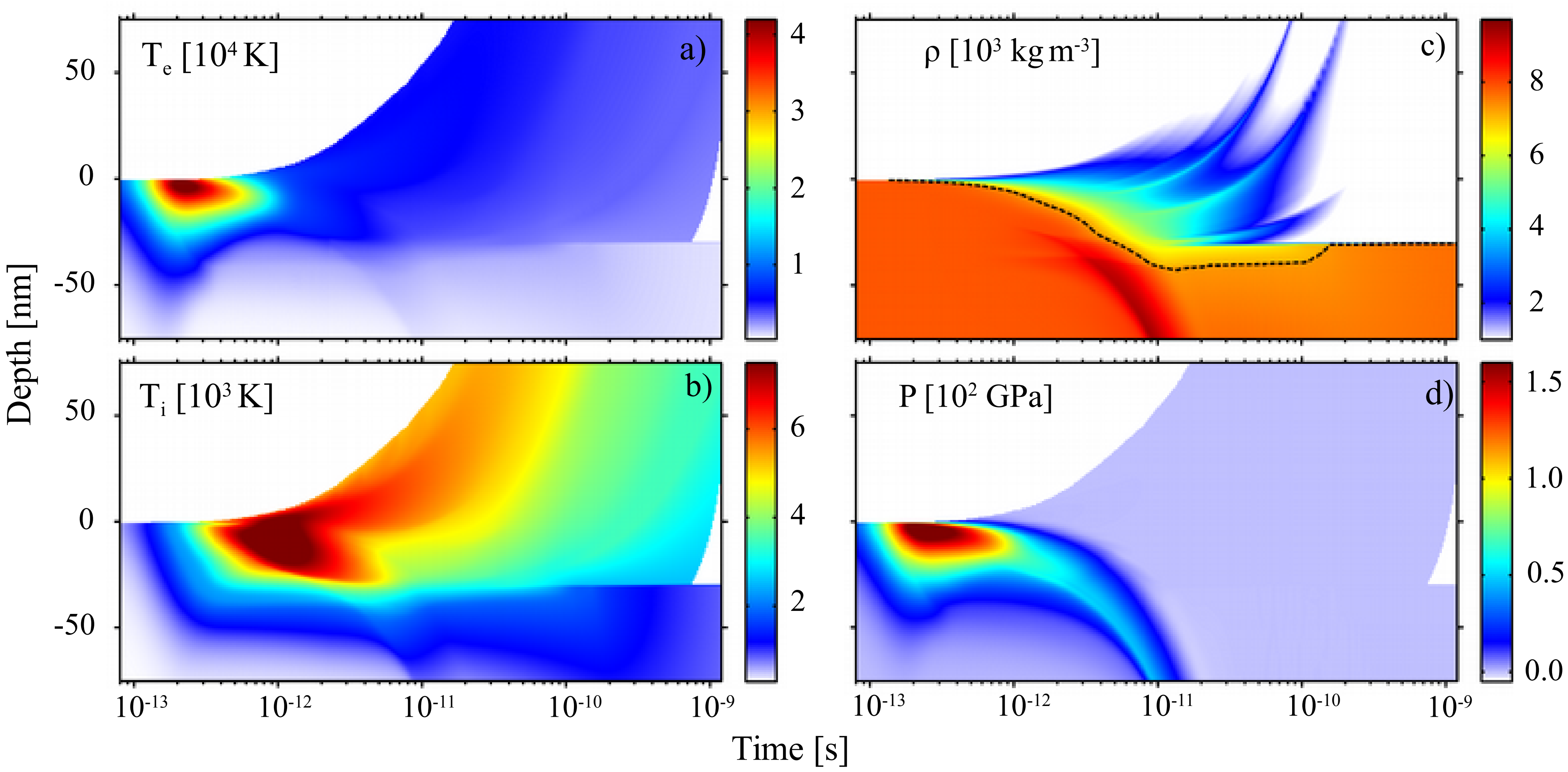
Based on DFPT calculations:

$$\lambda \langle \omega^2 \rangle = \int_0^\infty \omega \alpha^2 F(\Omega) d\omega$$

- *High G(T_e) at low temperature, decrease with T_e*

→ Nonequilibrium database for Stainless Steel

Hydrodynamic Properties of Irradiated Stainless Steel (Esther Code)



- *Fluence set to 0.6 J cm^{-2}*
- *Strong increase of T_e and P_e*
- *Electron-phonon coupling leading to 30 nm ablation depth*

Resume

<u>Tungsten:</u>

- Determination of nonequilibrium optical properties of Tungsten
- Change of optical indices leading to plasmonic switch in agreement with experimental observation
- Agreement between Interference model and experimental observations

<u>316L Stainless Steel:</u>

- Determination of thermodynamic and transport properties: nonequilibrium database
- Inserted in hydrodynamic codes showing matter ablation in agreement with experimental data

Thank you for attention